



A Statistical Theory on the Turbulent Diffusion of Gaussian Puffs.

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Publication date:
1982

Document Version
Publisher's PDF, also known as Version of record

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Citation (APA):
Mikkelsen, T. (1982). *A Statistical Theory on the Turbulent Diffusion of Gaussian Puffs*. Danmarks Tekniske Universitet, Risø Nationallaboratoriet for Bæredygtig Energi. Denmark. Forskningscenter Risoe. Risoe-R No. 475

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A STATISTICAL THEORY ON THE TURBULENT DIFFUSION
OF GAUSSIAN PUFFS

Torben Mikkelsen

Abstract. The relative diffusion of a one-dimensional Gaussian cloud of particles is related to a two-particle covariance function $R_{abs}(\xi_{ij}, \tau) = \overline{u(x_i(t))u(x_i(t-\tau)-\xi_{ij})}$ in a homogeneous and stationary field of turbulence. This two-particle covariance function expresses the velocity correlation between one particle (i) which at time t is in the position x_i , and another (j), which at the previous time $t-\tau$ is displaced the fixed distance ξ_{ij} relative to $x_i(t-\tau)$. For $\xi_{ij} = 0$, R_{abs} reduces to the Lagrangian covariance function of a single particle. On the other hand, setting the time lag τ equal to zero, R_{abs} becomes a pure Eulerian (fixed point) covariance function.

For diffusion times that are small compared to the Lagrangian integral time scale of the turbulence, simple expressions are derived for the growth of the standard deviation $\sigma(t)$ of the cloud by assuming that the wave number spectrum corresponding

(continued on next page)

October 1982

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to the Eulerian space covariance $R_{abs}(\xi_{ij}, 0)$ can be expressed as a power law function δk^p , where δ is a constant with dimension of $[\text{length}]^{(1+p)}$. For instance, by setting $p = -5/3$, an initially small cloud is found to grow as $\sigma^2(t) = (\overline{u^2})^{3/2} (2\Gamma(\frac{2}{3})\delta)^{3/2} t^3$ in agreement with Batchelor's (1950) inertial subrange theory. Correspondingly, for the enstrophy cascade subrange in two-dimensional turbulence, for which case $p = -3$, the theory yields $\sigma^2(t) = \sigma_0^2 \exp(\overline{u^2} \delta t^2)$, where σ_0 denotes the initial size of the cloud.

UDC 551.511

ISBN 87-550-0947-5

ISSN 0106-2840

Risø repro 1983

The present report (Risø-R-475) is part of the thesis:

FORMULATION AND EXPERIMENTAL EVALUATION OF AN OPERATIONAL
PUFF DIFFUSION MODEL

submitted to the Technical University of Denmark together with
the reports Risø-R-476 and Risø-R-479 in partial fulfilment of
the requirement for the degree of lic.techn. (Ph.D.).

Professor K. Refslund acted as responsible supervisor and
mag.scient. Leif Kristensen functioned as advisor. Professor
E. Eliassen was appointed external examiner.

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1. INTRODUCTION

Perhaps the most important property of turbulent fluid motion is its ability to disperse fluid particles which were initially close together. This property is of practical importance for the dispersal and dilution of pollutants in the environment and is also a general feature of the nature of turbulence.

The very first theories on eddy diffusion in the atmosphere put forward almost simultaneously by G.I. Taylor (1915) and L.F. Richardson (1922) were direct generalisations of the classical theory of molecular diffusion. They assumed that the transport properties of the eddies were entirely similar, except for a scale difference, to that of the molecules. Thus it was suggested that an eddy-diffusivity of the order 10^{-2} to $10^7 \text{ m}^2 \text{ s}^{-1}$ should replace a molecular diffusivity of the order $10^{-5} \text{ m}^2 \text{ s}^{-1}$ in entirely similar differential equations. It soon became clear, however, that the difference between the eddy structure of a turbulent fluid and the molecular structure of a fluid at rest was more than one of scale. The failure of this early theory became evident by the enormous variations found in the eddy diffusivities K . Richardson evaluated K for the diffusion of smoke over short distances, for the distribution of volcanic ash, and for the scatter of small balloons, and found K 's varying from 1 to $10^4 \text{ m}^2 \text{ s}^{-1}$. Other estimates varied from 10^{-2} to $10^7 \text{ m}^2 \text{ s}^{-1}$. In general, it was found that K increased rapidly with the scale of the phenomenon. The need of an extended theory to express the observed differences led G.I. Taylor (1921) to formulate the problem of diffusion by continuous movement. In his contribution to the subject, G.I. Taylor extended the theory on the problem of the scatter caused by uncorrelated movements in a fluid to the case where a correlation exists between the motion of a particle at one instant and its motion at some subsequent time. By doing so, Taylor solved the problem of relating single-particle dispersion in homogeneous turbulence to Lagrangian statistics of the velocity field.

The fundamentally different characteristics of two-particle statistics, or the statistics of a dispersing cloud of marked fluid in a turbulent field were first considered by F.L. Richardson (1926, 1929), and later by Batchelor (1950) and Brier (1950). Richardson (1926) pointed out that relative dispersion is an accelerating process in which an initially marked volume of fluid is spread at a rate depending on its size. Richardson (1926) summarized various atmospheric diffusion data (over the range of 1 to 10 km) and arrived at the "4/3-power law" for the relative, or instantaneous, diffusion coefficient K_R defined by

$$K_R = \alpha \ell^{4/3} \quad (1.1)$$

where ℓ is the distance separating two typical marked fluid elements and α is a constant. A notation list is contained in Appendix A.

To describe the shape characteristics of a dispersing cloud, F.L. Richardson (1926) introduced the distance neighbour function $q(\ell, t)$, which in a homogeneous and isotropic field can be defined by

$$q(\ell, t) = \frac{1}{Q} \int_{-\infty}^{\infty} C(\ell + \ell', t) C(\ell', t) d\ell' \quad (1.2)$$

where

$$Q = \int_{-\infty}^{\infty} C(\ell, t) d\ell$$

and $C(\ell, t)$ is the instantaneous concentration distribution along an arbitrarily oriented line ℓ at time t . The quantity $q(\ell, t)$ is an even function and its second and fourth moments are simply related to those of the concentration curve by

$$\sigma^2 = \frac{1}{2} \overline{\ell^2} \quad (1.3)$$

$$\mu = \frac{1}{2} \overline{\ell^4} - \frac{3}{4} (\overline{\ell^2})^2$$

where σ^2 and μ are the second and fourth moments of $C(\ell)$ about its centre of mass at a given time t .

Richardson also suggested the differential equation

$$\frac{\partial q}{\partial t} = \frac{\partial}{\partial \ell} \left(\alpha \ell^{4/3} \frac{\partial q}{\partial \ell} \right) \quad (1.4)$$

to describe the variable q . This has the solution (G.K. Batchelor, 1952)

$$q(\ell, t) = M \left(\frac{2}{3\pi^{1/2}} \right) \left(\frac{9}{4\alpha t} \right)^{3/2} \exp \left(- \frac{9\ell^{2/3}}{4\alpha t} \right), \quad (1.5)$$
$$\overline{\ell^2} = \frac{35}{9} \frac{2}{3} \alpha t^3$$

for the initial condition $q(\ell, 0) = M\delta(\ell)$, where $\delta(\ell)$ is the Dirac delta function, together with the constraint $\int_{-\infty}^{\infty} q(\ell, t) d\ell = M$. Note that the formulation by Richardson in Eq. (1.4) implies that the spreading of two marked fluid elements depends upon their instantaneous random separation ℓ .

A theoretical interpretation of the empirical relation Eq. (1.1) was later given by Obukhov (1941) and Batchelor (1950, 1952) in terms of the universal similarity theory of Kolmogorov. For the inertial subrange of high Reynolds number flow, Batchelor deduced that

$$K_R = c \epsilon^{1/3} \ell^{4/3} \quad (1.6)$$

where c is a constant of order unity and ϵ is the rate of energy dissipation.

The significance of introducing two-particle statistics in the relative dispersion problem was recognized by both Brier (1950) and Batchelor (1950) who independently demonstrated the involvement of the correlation between velocities of two different particles separated in both space and time. This two-particle Lagrangian correlation function is now well known to be fundamental

to the relative or cloud dispersion problem in the same way as the single-particle Lagrangian correlation function is fundamental to the absolute diffusion problem. Consider for simplicity a one-dimensional homogeneous and stationary turbulence with zero mean wind velocity. Following Batchelor (1950), the equation for the mean-square separation of an arbitrary pair of particles then becomes

$$\frac{d}{dt} \overline{\ell^2(t)} = 2 \int_0^t \overline{\{u_1(t) - u_2(t)\} \cdot \{u_1(\tau) - u_2(\tau)\}} d\tau, \quad (1.7)$$

where the subscripts identify the particles, u is the particle velocity, and overbars represent an ensemble average over a large number of realizations of the turbulent field, and t and τ are two different times.

Equation (1.7) contains two types of velocity product: The first, of the form $u_1(t) \cdot u_1(\tau)$ refers to the same particle at two different times and thus represents a Lagrangian single particle velocity covariance. The second, of the form $u_1(t) \cdot u_2(\tau)$ involves one particle at time t and a second at time τ and is thus a two-particle Lagrangian covariance at different instants.

An alternative to F.L. Richardson's formula (Eq. (1.4)) to describe the shape characteristics of a dispersing cloud was also given by Batchelor (1952), in which the effective diffusivity depends on the statistical quantity $\overline{\ell^2}$ rather than on the random instantaneous separation ℓ :

$$\frac{\partial \overline{q}}{\partial t} = \alpha (\overline{\ell^2})^{2/3} \frac{\partial^2 \overline{q}}{\partial \ell^2} \quad (1.8)$$

The solution satisfying the same conditions as Eq. (1.5) is here

$$\overline{q}(\ell, t) = \frac{1}{(2\pi \overline{\ell^2})^{1/2}} \exp\left(-\frac{1}{2} \frac{\ell^2}{\overline{\ell^2}}\right),$$

$$\overline{\ell^2}(t) = \left(\frac{2}{3} \alpha t\right)^3 \quad (1.9)$$

where \bar{q} denotes an ensemble-averaged value of the distance neighbour function, taken over concentration distributions arising from the release of a large number of identical clouds of marked fluid.

The form of the two solutions, Eqs. (1.5) and (1.9), are significantly different, and this large difference allowed Sullivan (1971) to test the two hypotheses against each other, using relatively crude, but repeated observations of dye plumes. His results showed that the average of several instantaneous concentration distributions about their centre of mass were approximately Gaussian and the ensemble-averaged distance-neighbour function to be of approximately Gaussian form. Thus the data were consistent with Eq. (1.9) and the theoretical approach of Batchelor (1952).

Various attempts to verify experimentally Batchelor's (1950) theory on the two-particle Lagrangian correlation function, Eq. (1.7) (Gifford, 1957a,b; 1977), have so far not thrown light on the nature of this function, or its effect on relative dispersion. Only qualitative agreement is found with Batchelor's inertial range theory for small times

$$\overline{\ell^2} = \overline{\ell_0^2} + 2(\overline{u_1(0)u_2(0)}) \cdot t^2 \quad (1.10)$$

and for intermediate times

$$\overline{\ell^2} = c \cdot \epsilon t^3 \quad (1.11)$$

where ℓ_0 is the initial separation of the pair of particles and c is a constant of order unity. However, various approximate forms of the two-particle Lagrangian correlation have been proposed (Brier 1950; Batchelor 1952; Smith and Hay 1961; C.J.P. Van Buijtenen 1982). Sawford (1982) compared the mean-square separation predictions from the first three of these and also from an approximation suggested by G.I. Taylor (see Batchelor 1952), in which the two-particle covariance for different instants is replaced by a simple product of a two-particle covariance at the same time and the single-particle Lagrangian auto-

covariance function, R_L . That is,

$$\begin{aligned}\overline{u_1(t) \cdot u_2(\tau)} &= \overline{u_1(t) \cdot u_2(t) \cdot R_L(t-\tau)} \\ &= \overline{u_1(t) \cdot u_2(t) \cdot u_1(\tau) \cdot u_1(t)} / \overline{u^2}\end{aligned}\quad (1.12)$$

By comparison with suitably documented observations, Sawford found this approximation to be the most appropriate.

In the next chapter the kinematics of particles involved in a relative diffusion process is discussed. In Chapter 3 the derivation of a formula for the growth rate of a one-dimensional Gaussian puff (or cloud) of particles is given. Finally, in Chapter 4 implications of the theory developed in Chapter 3 to various atmospheric dispersion problems are presented.

Throughout the rest of this report it will be assumed that the theory is restricted to scales large compared with the Kolmogorov scale $(\nu^3/\epsilon)^{1/4}$ (Batchelor 1950) so that the effects on molecular diffusion may be ignored.

2. THEORY

2.1. Dispersion in a frame of reference attached to the centre of mass

Consider the release of a cloud of marked fluid at the position $\underline{x} = 0$ and at time $t = 0$ into a field of stationary and homogeneous turbulence. Let the observed concentration field at subsequent times of the experiment be given by $C(\underline{x}, t)$. This field is subject to the continuity equation, which in integral form reads

$$Q = \int C(\underline{x}, t) d\underline{x} \quad (2.1)$$

The quantity Q is the total amount of matter released with the puff. The volume integral extends over all space. Based on the fundamental assumption about equivalence between ensemble-averaged concentration functions and probability distribution functions (see, e.g. Csanady, 1973), the quantity $Q^{-1} \overline{C(\underline{x}, t)} d\underline{x}$ describes the probability that a member of the marked fluid cloud will be found in the volume element $d\underline{x}$ surrounding the point \underline{x} , at time t . The first moment of the instantaneous normalized concentration field $Q^{-1} C(\underline{x}, t)$ yields the instantaneous position $\underline{c}(t)$ of the centre of mass of the cloud

$$\underline{c}(t) = \frac{1}{Q} \int \underline{x} C(\underline{x}, t) d\underline{x} \quad (2.2)$$

Like any single "marked" fluid particle, $\underline{c}(t)$ executes random movements as a function of time in a turbulent environment. The velocity of the centre of mass position vector, $\underline{V}_{cm} = d\underline{c}/dt$, follows from a differentiation of Eq. (2.2). Using the continuity equation in differential form

$$\frac{\partial C}{\partial t} = -\nabla \cdot (\underline{u} C(\underline{x}, t)) , \quad (2.3)$$

where \underline{u} is the velocity vector of the fluid and $\nabla \cdot$ is the divergence operator, and requiring that the concentration is well behaved so that $\lim_{|\underline{x}| \rightarrow \infty} \underline{x} C(\underline{x}, t) = 0$, Eq. (2.2) becomes

$$\underline{V}_{cm}(t) = \frac{1}{Q} \int \underline{u} C(\underline{x}, t) d\underline{x} \quad (2.4)$$

A coordinate system \underline{y} , attached to the puff centre of mass \underline{c} , may now be defined by

$$\underline{y} = \underline{x} - \underline{c} \quad (2.5)$$

This "relative" or "moving" frame of reference is exposed to ceaseless accelerations by the turbulence and is as such a non-inertial reference frame.

The observed concentration field may as well be described in this "relative" frame. Clearly, $C(\underline{x}, t) = C(\underline{y} + \underline{c}, t)$. The relative frame description $\tilde{C}(\underline{y}, t)$ differs from the "fixed" frame description $C(\underline{x}, t)$ only in the trivial point of a different coordinate origin. However, as will be shown, significant differences exist between the statistical properties of C as observed at a fixed \underline{x} and fixed \underline{y} , respectively.

The ensemble average of the velocity of the centre-of-mass vector \underline{V}_{cm} may be determined from Eq. (2.4).

$$\overline{\underline{V}_{cm}} = \frac{1}{Q} \int (\underline{\bar{u}} \bar{C} + \overline{\underline{u}'C'}) d\underline{x} \quad (2.6)$$

Primes denote fluctuations, i.e. departures from the ensemble mean in an individual realization. The mean product $\overline{\underline{u}'C'}$ is identified as a local turbulent flux vector. Csanady (1973, p. 86) argues that for a homogeneous field and provided that the cloud when released is symmetrical about the origin, this flux must be antisymmetrical, so that its space-integral is zero. Thus, for symmetrically released clouds, and for others at least approximately, the relation

$$\overline{\underline{V}_{cm}} = \frac{1}{Q} \int \underline{\bar{u}} \bar{C}(\underline{x}, t) d\underline{x} \quad (2.7)$$

substitutes for Eq. (2.6).

In the homogeneous field of consideration, the mean velocity $\overline{\underline{V}_{cm}}$ of the diffusing cloud will equal that of the mean wind velocity. Without loss of generality, the 'fixed' coordinate \underline{x} can be allowed to drift with constant mean velocity $\underline{\bar{u}}$, i.e. the coordinate \underline{x} can be chosen so as to make $\overline{\underline{V}_{cm}}(t) = \underline{0}$. By assuming this, the zero'th and first moments of the cloud, calculated on the basis of the ensemble average over many realizations of the flow, becomes in the fixed (\underline{x}) and moving (\underline{y}) frames, respectively

$$Q = \int \bar{C}(\underline{x}, t) d\underline{x} = \int \tilde{C}(\underline{y}, t) d\underline{y} \quad (2.8)$$

$$\underline{\bar{c}} = \int \underline{x} \bar{C}(\underline{x}, t) d\underline{x} = \int \underline{y} \tilde{C}(\underline{y}, t) d\underline{y} = 0$$

Any physically meaningful difference between 'fixed' and 'moving' frame ensemble-averaged concentration fields $\bar{C}(\underline{x}, t)$ and $\tilde{C}(\underline{y}, t)$ are therefore related entirely to their second and higher moments.

The second moments of the concentration distribution in the \underline{x} and \underline{y} frames are also simply related. By use of the definition of the centre-of-mass Eq. (2.2), and by noting that $\tilde{C}(\underline{y}, t) = C(\underline{x}, t) d\underline{x}$ for $\underline{x} = \underline{c} + \underline{y}$, we have for each of the three Cartesian coordinate components*

$$\begin{aligned} & \int_{-\infty}^{\infty} y^2 \tilde{C}(\underline{y}, t) d\underline{y} \\ &= \int_{-\infty}^{\infty} (\underline{x} - \underline{c})(\underline{x} - \underline{c}) C(\underline{x}, t) d\underline{x} \\ &= \int_{-\infty}^{\infty} \underline{x}^2 C(\underline{x}, t) d\underline{x} + \underline{c}^2 \int_{-\infty}^{\infty} C(\underline{x}, t) d\underline{x} \\ & \quad - 2\underline{c} \int_{-\infty}^{\infty} \underline{x} C(\underline{x}, t) d\underline{x} \\ &= \int_{-\infty}^{\infty} \underline{x}^2 C(\underline{x}, t) d\underline{x} - \underline{c}^2 Q . \end{aligned} \quad (2.9)$$

By repeating a given release a large number of times, an ensemble-averaged value of Eq. (2.9) may be obtained. When thus ensemble

*(Where all the variables refer to the same Cartesian coordinate component, specific designations of the individual components (1,2,3) have been omitted for simplicity.)

averaged, the left-hand side of Eq. (2.9) may be identified as the mean-square spread of the cloud, calculated in the moving frame of reference, y

$$\overline{y^2}(t) = \frac{1}{Q} \int_{-\infty}^{\infty} y^2 \bar{C}(y,t) dy, \quad (2.10)$$

whereas the first term on the right-hand side may be identified as the mean-square spread of the particles in the 'fixed' frame of reference, x

$$\overline{x^2}(t) = \frac{1}{Q} \int_{-\infty}^{\infty} x^2 \bar{C}(x,t) dx \quad (2.11)$$

The last term, $\overline{c^2}(t)$ represents the mean-square spread of the "centre-of-mass" movement of the puffs, also referred to the fixed frame of reference.

Eq. (2.9) can now be written as

$$\overline{x^2}(t) = \overline{y^2}(t) + \overline{c^2}(t) \quad (2.12)$$

which states that the spread, referred to an absolute frame of reference, of an ensemble of clouds which are released at $\underline{x} = 0$, equals at time $t > 0$ the sum of the relative spread of the puff and the spread of the centre-of-mass movement of the puff referred to the absolute frame of reference. Clearly, $\overline{x^2}$ is always greater than either $\overline{y^2}$ or $\overline{c^2}$.

In the previous section it was mentioned that relative diffusion is closely related to the rate at which two arbitrary diffusing particles separate (cf. the discussion in connection with Eq. (1.7)). To establish a relationship between the mean-square separation $\overline{\ell^2}$ (Eq. (1.7)) of two diffusing particles belonging to the cloud and the mean-square distance from the centre-of-gravity $\overline{y^2}$ (Eq. (2.10)), let an infinitely small cloud be released at $t = 0$ at the origin of a fixed coordinate x , and consider the mean product

$$1/Q^2 \overline{C(x,t) C(x',t)} dx dx' \quad (2.13)$$

As discussed earlier, the quantity $Q^{-1} \overline{C(x,t)}$ is equal to the probability that a marked fluid will be found in the small interval between x and $x+dx$, at successive times t . This is also equal to the probability of displacement x in time t for a single diffusing particle. The product may be regarded as the joint probability of finding marked fluid particles both at x and x' , hence it is also equal to the joint probability of particle displacements for two diffusing particles x and x' , in the time period t . Denoting the two-particle displacement probability density by $P(x,x',t)$, such that $P(x,x',t) dx' dx$ is the probability of finding one particle at x , and another at x' , we may also write

$$\overline{C(x,t) C(x',t)} = Q^2 P(x',t) \quad (2.14)$$

The second moment of $P(x,x',t)$ with respect to the separation $(x-x')$ yields the mean-square separation $\overline{\ell^2}$ of two diffusing particles, along x

$$\begin{aligned} \overline{\ell^2} &= \iint (x'-x)^2 P(x,x',t) dx' dx \\ &= \frac{1}{Q^2} \iint (x'-x)^2 \overline{C(x,t) C(x',t)} dx' dx \\ &= 2 \overline{x^2(t)} - 2 \overline{c^2(t)} \end{aligned} \quad (2.15)$$

By use of Eq. (2.12) this simply becomes

$$\overline{\ell^2} = 2 \overline{y^2(t)} \quad (2.16)$$

which states that the mean-square separation of two diffusing particles along an arbitrary coordinate direction is just twice their mean-square separation from the centre of mass.

The probability density $P(x,x',t)$ may alternatively be regarded as specifying the probability of an absolute displacement x ,

and a relative displacement $\xi = x' - x$ of the two particles. Multiplying P by Q and integrating over all displacement x yields the ensemble mean of the distance neighbour function mentioned in the previous paragraph:

$$\begin{aligned}\bar{q}(\xi, t) &= Q \int P(x, x', t) dx \\ &= \frac{1}{Q} \int \bar{C}(x, t) \bar{C}(x + \xi, t) dx\end{aligned}\quad (2.17)$$

The integral over the concentration product can be determined for individual realizations and yields a somewhat smoothed picture of the distribution of particles within the cloud. As suggested by F.L. Richardson (1926), this ensemble-averaged neighbour density constitutes a possible description of relative diffusion alternative to the mean concentration distribution in a moving frame (cf. Eq. (1.2)).

2.2. Kinematics of particle movements in a moving frame

Let the velocities of the marked fluid or suspended particles referred to the moving frame be $\underline{v}(v_1, v_2, v_3)$. By differentiation with time of Eq. (2.5) it then follows that

$$\underline{v} = \underline{u} - \underline{V}_{cm} \quad (2.18)$$

From Eq. (2.8) we have $d\bar{C}/dt = \bar{V}_{cm} = 0$, and without loss of generality we may assume that $\bar{\underline{u}} = 0$ (by measuring \underline{u} relative to a frame of reference moving with any mean motion of the ensemble).

Then, from Eq. (2.18) it is also clear that the ensemble-averaged velocity of a particle, relative to the centre-of-mass coordinate of the cloud is zero.

$$\bar{\underline{v}} = d\bar{\underline{y}}/dt = 0 \quad (2.19)$$

Because the relative velocity and displacement of the diffusing particles within the puff are related by the Lagrangian integral,

$$\underline{y} = \int_0^t \underline{v}(t') dt' \quad (2.20)$$

an analogue to Taylor's theorem, using relative velocities, can formally be derived. Along individual Cartesian coordinate directions, the mean-square displacement of the cloud varies as

$$\frac{d\overline{\langle y^2 \rangle}}{dt} = 2\overline{\langle y \frac{dy}{dt} \rangle} = 2 \int_0^t \overline{\langle v(t)v(t') \rangle} dt' \quad (2.21)$$

where v is the component of the Lagrangian velocity vector \underline{v} that is parallel to y .

Two types of averaging are involved here. As previously, the overbars indicate ensemble averaging over all realizations of the turbulent field whereas the brackets $\langle \rangle$ imply an average over all marked fluid or particles in a particular cloud. It must also be emphasized, however, that the relative velocity $v(t)$, in contrast to the absolute velocity $u(t)$ usually used with Taylor's theorem, does not constitute a stationary process. At the beginning when an initially small cloud is released, only the smallest turbulent eddies contribute to $v(t)$ and thereby to the growth, then increasingly larger ones, until the maximum eddy size is reached and exceeded. The velocity covariance $\overline{\langle v(t)v(t') \rangle}$ is thus not only a function of time lag $\tau = t-t'$, but depends also on the diffusion time t explicitly.

A modified Lagrangian correlation function can formally be introduced which is appropriate for the relative velocities of particles within the cluster (Csanady, 1970)

$$r(t, \tau) = \frac{\overline{\langle v(t)v(t-\tau) \rangle}}{\overline{\langle v^2(t) \rangle}} \quad (2.22)$$

The qualitative behaviour of this relative velocity correlation function is shown in Fig. 1.

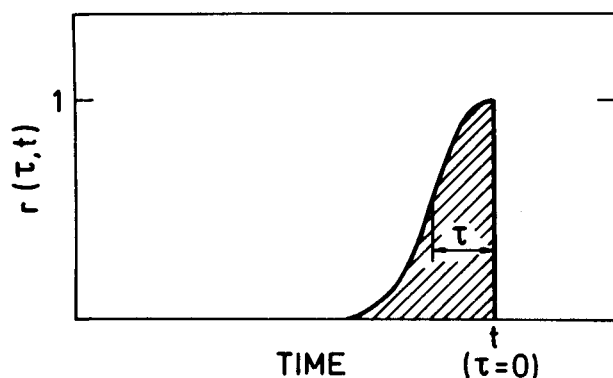


Fig. 1. Qualitative behaviour of relative velocity correlation $r(t, \tau)$ for given t . The shaded area visualizes the relative integral time scale $t_r(t)$.

At zero time lag $\tau = 0$, $r(t, \tau)$ has its maximum value of unity. As with the Lagrangian correlation functions of absolute velocities (for instance, discussed by Tennekes and Lumley (1972)), $r(t, \tau)$ probably remains a monotonically decreasing function of the time lag τ . Formally, $r(t, \tau)$ defines a Lagrangian integral time scale $t_r(t)$ appropriate for relative diffusion, which can be visualized as the shaded area in Fig. 1:

$$t_r(t) = \int_0^t r(t, \tau) d\tau \quad (2.23)$$

Here the time of release of the cloud is arbitrarily set equal to zero in the lower limit of the integral, and consequently the time lag τ is confined to lie within the interval between zero and t .

This relative Lagrangian time scale is characteristic for the average lifetime of eddies contributing to the movement of the particles relative to the centroid of the cloud. These eddies range from a size comparable to the size of the cloud down to the smallest length scale of the fluid, i.e. the Kolmogorov scale $(\nu^3/\epsilon)^{1/4}$. As will be discussed in the chapter to follow, the eddies of a size comparable to the dimension of the cloud are those that most efficiently contribute to cloud dispersion. This is true at least for diffusion in ranges where the energy spectrum is a decreasing function of the wavenumber.

In this region the time scale $t_r(t)$ must be expected to be closely related to the decay time of eddies of size comparable

to that of the cloud. A simple estimate of $t_r(t)$ is $(\overline{y^2}/\overline{v^2})^{1/2}$. As the cloud grows, successively larger eddies begin to contribute; the larger the eddy, the longer is its "memory" or decay time. From this qualitative argument it is understandable that t_r , and the mean-square relative velocity $\overline{v^2}$ as well, must be an increasing functions of the diffusion time t . Since $r(\tau, t)$ has the maximum value of unity and is a decreasing function for $\tau > 0$, an upper bound for the relative time scale is given by $t_r(\tau) \leq t$. Ultimately, when the cloud becomes so large that the particles associated with it move independently of each other for all practical purposes, t_r ceases to grow and becomes equal to the Lagrangian time scale of the fluid t_L . In this far field limit, $\overline{v^2}$ will also cease to grow and asymptotically approaches the variance of the fluid, $\overline{u^2}$.

By combining Eqs. (2.22) and (2.23), the second moment of the distribution function Eq. (2.21) may now be written as

$$\overline{y^2} = 2 \int_0^t \overline{v^2(t')} t_r(t') dt' \quad (2.24)$$

The equation represents a kinematic formulation of the relative mean-square spread defined in Eq. (2.10).

3. TURBULENT DIFFUSION OF GAUSSIAN PUFFS

3.1. Relative diffusion equation

Here the dispersion of passive one-dimensional clouds or puffs will be considered, released from an instantaneous point source in a homogeneous and stationary field of turbulence. In accordance with common practice, the particle density distribution function will be assumed to be Gaussian, and the growth of the cloud will be calculated in terms of the standard deviation σ of the density distribution function. By considering the cloud

dispersion to take place along a single but arbitrarily oriented direction in a Cartesian coordinate system, the analysis allows for calculating relative diffusion in situations where the turbulent field is not necessarily isotropic. This is of great practical importance. In the planetary boundary layer of the atmosphere, for instance, the turbulent field in the two horizontal component directions may be considered homogeneous, under certain conditions, but due to the presence of the ground, it may not be isotropic on scales where the relative diffusion of pollutants is of interest.

Chapter 2 led to a general kinematic formulation of the process of the relative diffusion of a cloud in the coordinate system moving with the centroid of the cloud. Here the starting point will be differential equation (2.21), which applies as well to the calculation of the growth of a one-dimensional Gaussian puff, the standard deviation of which is denoted by $(\overline{y^2})^{1/2} = \sigma(t)$

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \int_0^t \overline{v(t)v(t-\tau)} d\tau \quad (3.1)$$

As before, the particle velocity $v(t) = dy(t)/dt$ and the brackets refer to an average over all the particles in the cloud, which is assumed here to have a Gaussian density distribution. The over-bar indicates an ensemble averaging over the turbulent velocity field in question. By use of Eq. (2.18), the moving frame velocity covariance in Eq. (3.1) may next be related to the fixed frame particle velocity u and the velocity of the centroid V_{cm} .

$$\begin{aligned} \overline{v(t)v(t-\tau)} &= \overline{\{u(t) - V_{cm}(t)\} \{u(t-\tau) - V_{cm}(t-\tau)\}} \\ &= \overline{u(t)u(t-\tau)} - \overline{u(t)V_{cm}(t-\tau)} - \overline{u(t-\tau)V_{cm}(t)} \\ &\quad + \overline{V_{cm}(t)V_{cm}(t-\tau)} \end{aligned} \quad (3.2)$$

For convenience it is feasible to consider the Gaussian cloud as made up of a very large, but finite number N of individual particles. In this case the averaging over the particles in

the cloud $\langle \rangle$ becomes the operation $1/N \sum_{i=1}^N$. The subsequent transformation back to the continuous particle distribution function can then be achieved by letting N approach infinity. A reduction of the terms in Eq. (3.2) now follows from the fact that in homogeneous and stationary turbulence, Lagrangian auto-covariance functions of the individual and simultaneously released particles are identical. In the fixed frame, the i 'th particle's auto-covariance function reads $\overline{u_i(t)u_i(t-\tau)}$, where the suffix i refers to the i 'th particle of the cloud. In the moving frame, the same particle's auto-covariance function reads $\overline{v_i(t)v_i(t-\tau)}$.

The terms in Eq. (3.2) thereby become

$$\begin{aligned}\overline{\langle v(t)v(t-\tau) \rangle} &= \frac{1}{N} \sum_{i=1}^N \overline{v_i(t)v_i(t-\tau)} = \overline{v(t)v(t-\tau)} \\ \overline{\langle u(t)u(t-\tau) \rangle} &= \frac{1}{N} \sum_{i=1}^N \overline{u_i(t)u_i(t-\tau)} = \overline{u(t)u(t-\tau)} \\ \overline{\langle u(t)V_{cm}(t-\tau) \rangle} &= \overline{V_{cm}(t-\tau) \frac{1}{N} \sum_{i=1}^N u_i(t)} = \overline{V_{cm}(t-\tau)V_{cm}(t)} \\ \overline{\langle u(t-\tau)V_{cm}(t) \rangle} &= \overline{V_{cm}(t) \frac{1}{N} \sum_{i=1}^N u_i(t-\tau)} = \overline{V_{cm}(t)V_{cm}(t-\tau)}\end{aligned}\tag{3.3}$$

The first two of these equations states that the cloud-averaged ($\langle \rangle$) auto-covariance function, in moving and fixed coordinates respectively, equals the auto-covariance function of an individual particle. The quantity $1/N \sum_{i=1}^N u_i$ is analogous to the definition of the centre-of-mass velocity in Eq. (2.4).

By use of this, Eq. (3.2) now takes the simple form

$$\overline{v(t)v(t-\tau)} = \overline{u(t)u(t-\tau)} - \overline{V_{cm}(t)V_{cm}(t-\tau)}\tag{3.4}$$

Since the turbulence is assumed to be stationary, the Lagrangian auto-covariance $\overline{u(t)u(t-\tau)}$ must be independent of time t . This, however, is neither the case for the relative velocity covariance, nor for the centre-of-mass velocity covariance function in Eq. (3.4).

Setting $\tau = 0$, Eq. (3.4) reduces to

$$\overline{u^2} = \overline{v^2(t)} + \overline{V_{cm}^2(t)}, \quad (3.5)$$

where the right-hand side is explicitly written as functions of time in order to emphasize the non-stationarity of the terms. The equation states that the velocity variance of a particle or fluid element, measured in the fixed frame of reference $\overline{u^2}$, is partitioned in a complementary manner between the variance of the velocity of the centre of mass of the cloud, and the variance of velocities relative to this, $\overline{v^2}$. The same result is more easily derived by ensemble-averaging the square of Eq. (2.18) and making use of the zero value of $\langle v(t)V_{cm}(t) \rangle$ in the moving coordinate system.

An analogous Taylor's theorem, expressed in terms of relative coordinates and velocities was previously formulated in connection with Eq. (2.21). This theorem was originally derived (by G.I. Taylor, 1921) to describe the spreading $\overline{x^2}$ of individually released particles in a fixed frame of reference, but it applies as well to the spreading of the cloud's centre-of-mass coordinate $\overline{c^2}$. Therefore, the following set of equations describes the spreading along the Cartesian coordinate direction considered:

$$\begin{aligned} \frac{1}{2} \frac{d\overline{x^2}}{dt} &= \int_0^t R_{abs}(\tau) d\tau ; & R_{abs}(\tau) &\equiv \overline{u(t)u(t-\tau)} \\ \frac{1}{2} \frac{d\overline{c^2}}{dt} &= \int_0^t R_{cm}(t, \tau) d\tau ; & R_{cm}(t, \tau) &\equiv \overline{V_{cm}(t)V_{cm}(t-\tau)} \\ \frac{1}{2} \frac{d\overline{y^2}}{dt} &= \int_0^t R_{rel}(t, \tau) d\tau ; & R_{rel}(t, \tau) &\equiv \overline{v(t)v(t-\tau)} \end{aligned} \quad (3.6)$$

In Eq. (3.6), the Lagrangian covariance functions for the (absolute) velocity in the fixed frame x , for the velocity of the centre-of-mass coordinate c , and for the (relative) velocity in the moving frame y , have been abbreviated by $R_{abs}(\tau)$ ^{*}, $R_{cm}(t, \tau)$ and $R_{rel}(t, \tau)$, respectively.

By substituting the first of the Eqs. (3.3) into Eq. (3.1), and by subsequent use of Eq. (3.6), the following relation is easily obtained:

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \frac{1}{2} \frac{d\overline{x^2}}{dt} - \frac{1}{2} \frac{d\overline{c^2}}{dt} \quad (3.7)$$

When integrated with respect to the time t , this equation becomes identical to the previous finding in Eq. (2.10).

In contrast to Eq. (2.10), however, the present equation constitutes a foundation on which the appropriate velocity covariance functions can be included to give the rate of growth of the cloud. A combination of Eqs. (3.6) and (3.7) gives

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \int_0^t \{R_{abs}(\tau) - R_{cm}(t, \tau)\} d\tau \quad (3.8)$$

Equation (3.7), with $\sigma^2 = \frac{1}{2} \overline{\ell^2}$ (where $\overline{\ell^2}$ is the mean-square separation of the particles) compares with the general formulation of the relative diffusion concept originally presented by Batchelor (1952) but also with Sawford (1982) (cf. Eq. (3) of the latter article).

In Eq. (3.8), $R_{abs}(\tau)$ denotes the Lagrangian covariance function appropriate for single-particle diffusion. In order to be able to integrate Eq. (3.8), however, $R_{cm}(t, \tau)$ must also be related

^{*}) To emphasize independence of the absolute time t , R_{abs} is defined here as a function of the time-lag τ alone.

to some fundamental statistical property of the turbulence. An attempt to do so is suggested in the following:

The centre-of-mass auto-covariance function is by definition given by

$$R_{cm}(t, \tau) = \overline{V_{cm}(t)V_{cm}(t-\tau)} = \overline{\langle u(t) \rangle \langle u(t-\tau) \rangle} \quad (3.9)$$

As previously discussed, the brackets in Eq. (3.9) symbolize an (instantaneous) average over all the individual particles or marked fluid in the cloud. As shown above, this average can be arrived at by use of the instantaneous displacement distribution function of the cloud which, when referred to the fixed coordinate x , reads $Q^{-1} C(x, t)$. When multiplied by the (large) number N of particles that constitute the cloud, $Q^{-1} C(x, t) dx$ denotes the (small) number of particles that occupy the position at time t between x and $x + dx$. At two fixed times, t and $t-\tau$, the expressions for the velocity of the cloud centroid as given by Eq. (2.4) therefore read, respectively

$$V_{cm}(t) = \frac{1}{Q} \int_{-\infty}^{\infty} u(x', t) C(x', t) dx' \quad (3.10)$$

$$V_{cm}(t-\tau) = \frac{1}{Q} \int_{-\infty}^{\infty} u(x'', t-\tau) C(x'', t-\tau) dx''$$

and with these relations, the centre-of-mass covariance function in Eq. (3.9) becomes

$$R_{cm}(t, \tau) = \frac{1}{Q^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{u(x', t) u(x'', t-\tau) C(x', t) C(x'', t-\tau)} dx' dx'' \quad (3.11)$$

From the very beginning of the present chapter it has been assumed that the form of the instantaneous displacement distribution function of the cloud $Q^{-1} C(x, t)$ develops in a similar way as a function of time. In accordance with general practice, this distribution was taken to be Gaussian and thereby normally dis-

tributed around the centroid $c(t)$ of the cloud, with a standard deviation $\sigma(t)$,

$$Q^{-1} C(x,t) = \frac{1}{\sqrt{2\pi} \sigma(t)} \exp\left\{-\frac{1}{2} (x-c(t))^2/\sigma^2(t)\right\} \quad (3.12)$$

This equation therefore constitutes a fundamental assumption on which the present theory depends. We shall return to this point in the subsequent discussion. When inserting this in Eq. (3.11), however, the averaging over the turbulent field represented by the overbar still has to extend over the displacement distribution functions because the centroid $c(t)$ moves around in a random manner as a function of time. But by use of the substitution $x = c+y$, the frame of reference can be changed from fixed (x) to moving (y) coordinates. In the moving frame, the Gaussian particle density distribution function, $G_{\sigma(t)}(y,t)$ becomes

$$G_{\sigma(t)}(y,t) = Q^{-1} C(c+y,t) = \frac{1}{\sqrt{2\pi} \sigma(t)} \exp\left\{-\frac{1}{2} y^2/\sigma^2(t)\right\} \quad (3.13)$$

In addition, the following relation

$$Q^{-1} C(x,t) dx = G_{\sigma(t)}(y,t) dy \quad (3.14)$$

expresses the fact that the number of particles in a small line element is not influenced by changing from a fixed to moving frame of reference.

The velocity of the centroid corresponding to Eq. (3.10) now becomes, with the moving coordinate y as independent variable

$$V_{cm}(t) = \int_{-\infty}^{\infty} u(y'+c,t) G_{\sigma(t)}(y',t) dy' \quad (3.15)$$

$$V_{cm}(t-\tau) = \int_{-\infty}^{\infty} u(y''+c,t-\tau) G_{\sigma(t-\tau)}(y'',t-\tau) dy''$$

and analogous to Eq. (3.11), the centre-of-mass covariance function now becomes

$$R_{cm}(t, \tau) =$$

$$\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{u(y'+c, t) u(y''+c, t-\tau)} G_{\sigma}(t)(y', t) G_{\sigma}(t-\tau)(y'', t-\tau) dy' dy'' \quad (3.16)$$

As a consequence of the change of the reference frame the stochastic variable c is removed from the distribution functions and the averaging over the turbulent field therefore now affects only the velocity covariance $\overline{u(y'+c, t) u(y''+c, t-\tau)}$, as Eq. (3.16) shows. By changing the reference frame for the velocities to the moving coordinate as well, this covariance can be written as $\tilde{u}(y', t) \tilde{u}(y'', t-\tau)$, where $\tilde{u}(y, t) = u(y+c, t) = u(x, t)$. This covariance function will now be the subject of further investigation. It expresses an ensemble-averaged (generalized Eulerian) correlation of fluid velocity, measured at the two fixed points y' and y'' in the moving coordinate system at the two times t and $t-\tau$, respectively. The situation is shown in Fig. 2.

With the purpose of relating this fixed point velocity covariance to some more fundamental property of the turbulent flow, however, the underlying Lagrangian diffusion process of the problem has to be investigated.

Figure 3 shows the Lagrangian trajectory $y_i(t)$ of a particle or marked fluid (i) that at the previous time $t-\tau$ was in the position $y_i(t-\tau)$. In the moving frame, the displacement $\Delta y_i = y_i(t) - y_i(t-\tau)$ constitutes a stochastic process, having a continuous density distribution function G_S as indicated. The quantity $\overline{\Delta y_i^2}$ equals the i 'th particle's contribution to the growth of the cloud in the period of time between $t-\tau$ and t . The growth of the cloud in the period between $t-\tau$ and t is therefore the collective result of the motion of all the particle motion over that time interval. Taking the distribution function for the individual particles as identical and independent Gaussians, G_S , will now be shown to be consistent with the Gaussian distribution function $G_{\sigma}(t)$ assumed for the particle density of the cloud.

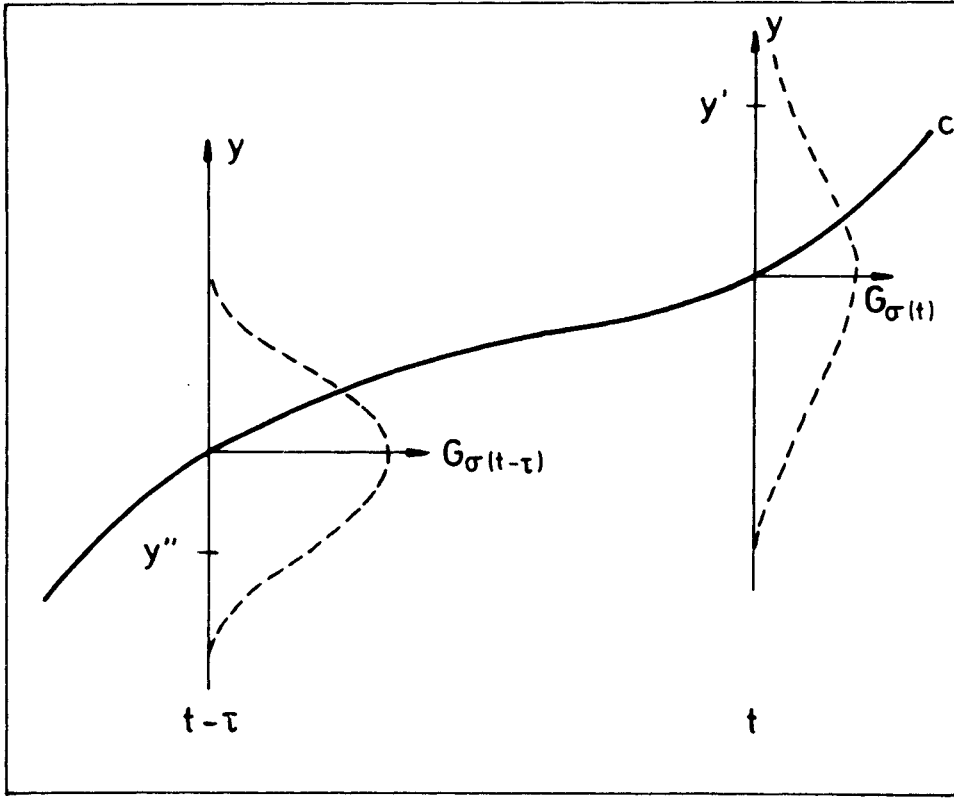


Fig. 2. The motion of a Gaussian cloud G in the fixed frame of reference x as a function of time t . The centre-of-mass coordinate of the cloud c defines the origin of the moving frame y , relative to which the dispersion of the cloud in terms of the standard deviation σ is defined. Also shown are the two fixed points in the moving frame, y' and y'' , on which the covariance function $\overline{u(y'+c,t)u(y''+c,t-\tau)}$ depends.

The independence of any neighbour particles of the distribution function G_S implies that $\overline{\Delta y_i \Delta y_j} = 0$ for $i \neq j$. Therefore, the distribution function $G_\sigma(t)$ can be calculated as a superposition of the dispersion from all the marked particles constituting the cloud. With the continuous distribution functions in question, this superposition leads to the integral (Mikkelsen et al. (1982))

$$G_\sigma(t)(y,t) = \int_{-\infty}^{\infty} G_S(y-y_0,t) G_\sigma(t-\tau)(y_0,t-\tau) dy_0 \quad (3.17)$$

With $G_\sigma(t)$ and $G_\sigma(t-\tau)$ inserted as Gaussian distributions having standard deviations equal to $\sigma(t)$ and $\sigma(t-\tau)$, respectively, G_S

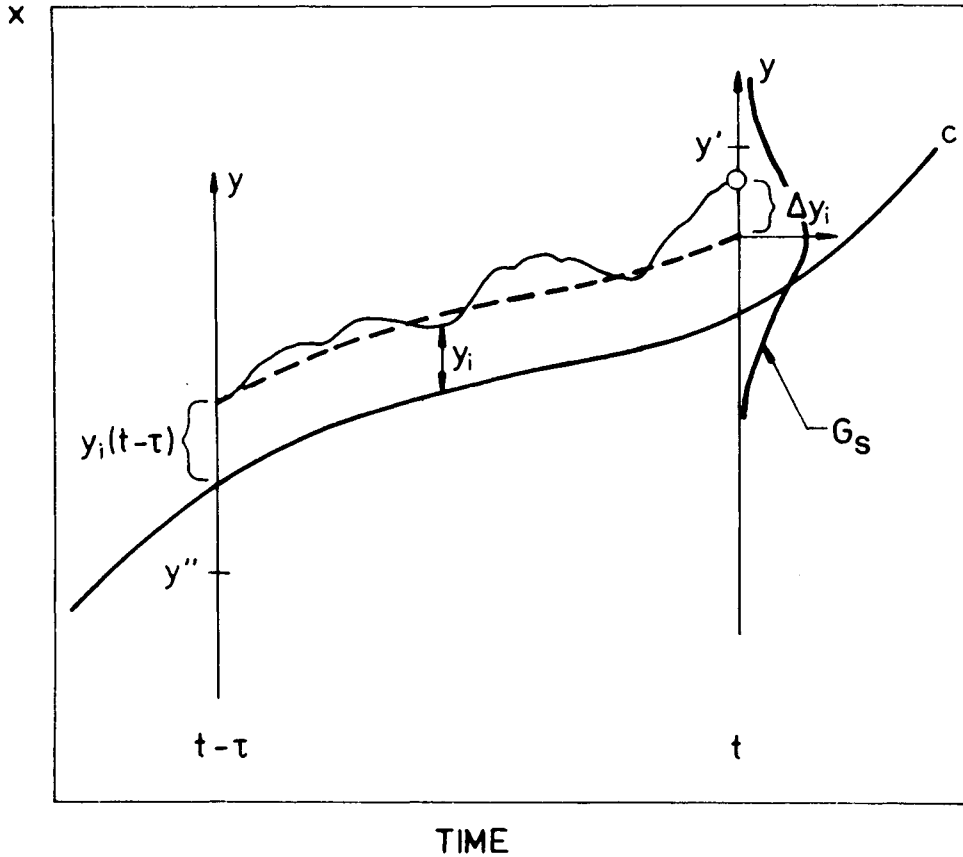


Fig. 3. The moving frame trajectory y_i of a marked fluid particle (i) that at time $t-\tau$ holds the position $y_i(t-\tau)$. The quantity $\Delta y_i \equiv y_i(t) - y_i(t-\tau)$ as well as its distribution function G_s is shown at time t .

can be solved by a Fourier transform of the integral equation (3.17). This is another Gaussian having a standard deviation squared given by

$$s^2 = \sigma^2(t) - \sigma^2(t-\tau) \quad (3.18)$$

Instead of assuming a priori that the instantaneous cloud density distribution $G_\sigma(t)$ is Gaussian, it could alternatively have been assumed that the individual particles' displacement distribution functions G_s in the moving frame are identical and independent Gaussians, with a standard deviation as given by Eq. (3.18). From Eq. (3.17) it then follows that an initial Gaussian distributed cloud, with standard deviation $\sigma(t-\tau)$, would remain Gaussian at all subsequent times with standard deviation $\sigma(t)$.

It can be claimed that the Gaussian property of the relative displacement process Δy_i , together with the relation

$$\overline{\Delta y_i \Delta y_j} = \begin{cases} \sigma^2(t) - \sigma^2(t-\tau) & \text{for } i = j \\ 0 & \text{for } i \neq j, \end{cases} \quad (3.19)$$

is the fundamental assumption of the present theory, and that a Gaussian cloud results as a consequence thereof.

The requirement that any two particles disperse uncorrelated in the moving frame (i.e. $\overline{\Delta y_i \Delta y_j} = 0$ for $i \neq j$), no matter how close they are, appears to be rather restrictive in a realistic turbulent field. When the cloud consists of a very large number of particles, however, in which case a continuum description of the turbulence applies, the requirement corresponding to $\overline{\Delta y_i \Delta y_j} = 0$ for $i \neq j$ is

$$l_r(t) \ll \sigma(t) \quad (3.20)$$

Here, l_r is the relative integral length scale of the turbulence, which in terms of the relative velocity v can be defined as

$$l_r(t) = \left(\overline{v^2(t)} \right)^{-1} \int_0^\infty \overline{v(y,t)v(y+\xi,t)} d\xi \quad (3.21)$$

The inequality $l_r \ll \sigma$ expresses the rather strong limitations that have to be put on the turbulent field in case the instantaneous particle distribution function of an initial Gaussian cloud is required to evolve in a Gaussian manner at all subsequent times. However, this requirement is not as restrictive as the corresponding two-particle requirement described by Eq. (3.19), especially when the cloud becomes large. With this picture in mind of the relative diffusion process, it is now possible to continue the calculation of the velocity covariance $\overline{u(y',t)u(y'',t-\tau)}$ in Eq. (3.16).

In close analogy to the turbulent dispersion of contaminant particles, the turbulent field itself can also be considered as consisting of a very large, but numerable number M of small fluid elements or fluid particles.

Suppose that the i 'th of these fluid particles is in position $y_i = y'$ at time t . The i 'th particle Lagrangian velocity $\tilde{u}(y_i(t))$ will then equal the Eulerian velocity $\tilde{u}(y', t)$ at this point and that time. Equivalently, if the j 'th fluid particle at time $t-\tau$ is in the position $y_j = y''$, its Lagrangian velocity equals the Eulerian velocity of that point, i.e. $\tilde{u}(y_j(t-\tau)) = \tilde{u}(y'', t-\tau)$.

Now consider the situation in Fig. 4 which shows the trajectories of an arbitrary chosen pair of fluid particles (i) and (j), the separation of which at time $t-\tau$ is given by ξ_{ij} , both in the fixed (x) and moving (y) frames as well

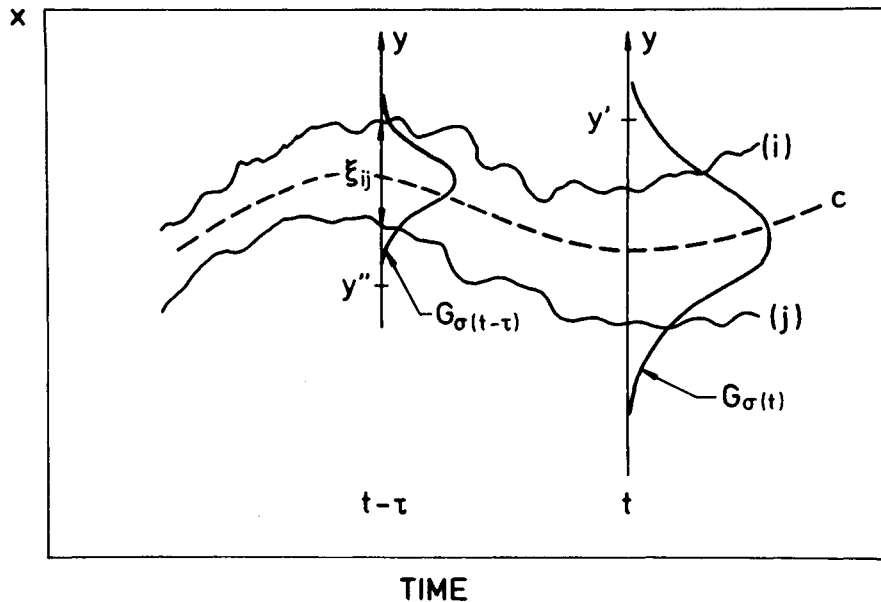


Fig. 4. The trajectory of an arbitrary fluid particle (j), which at time $t-\tau$ is in the position $y_j(t-\tau)$ and another particle (i), which at the same time holds a position displaced the distance ξ_{ij} relative to (j). Note that ξ_{ij} denotes the separation of the two particles in both the moving and fixed frames: $\xi_{ij} = y_i(t-\tau) - y_j(t-\tau) = x_i(t-\tau) - x_j(t-\tau)$. (Otherwise as in Fig. 3.)

$$\begin{aligned}
 \xi_{ij} &= x_i(t-\tau) - x_j(t-\tau) \\
 &= y_i(t-\tau) + c(t-\tau) - (y_j(t-\tau) + c(t-\tau)) \\
 &= y_i(t-\tau) - y_j(t-\tau)
 \end{aligned} \tag{3.22}$$

By fixing the variable ξ_{ij} at a constant value here, we can consider the joint probability distribution of the remaining two variables $y_i(t)$ and $y_j(t-\tau)$. This is called the conditional probability of $y_i(t)$, $y_j(t-\tau)$; it is conditional on ξ_{ij} having a prescribed value. It will be denoted by $S(y_i(t), y_j(t-\tau) | \xi_{ij})$.

Suppose one knew the conditional joint probability $S(y', y'' | \xi_{ij})$ for finding the fluid particle (i) in the position y' at time t and the fluid particle (j) in the position y'' at time $t-\tau$, under the condition that the separation of the two particles, at time $t-\tau$, is given by the fixed distance $\xi_{ij} = y_i(t-\tau) - y_j(t-\tau)$.

The contribution from this particular particle pair (i) and (j) to the total covariance $\overline{u(y', t)u(y'', t-\tau)}$ could then be calculated as $S(y', y'' | \xi_{ij}) \overline{u(y_i(t))u(y_j(t-\tau))}$, where the ensemble-averaged covariance function of the velocity of the pair

$$\begin{aligned}
 &\overline{u(y_i(t))u(y_j(t-\tau))} \\
 &= \overline{u(y_i(t))u(y_i(t-\tau) - \xi_{ij})}
 \end{aligned} \tag{3.23}$$

is also subject to the condition that the particle pair separation at time $t-\tau$ equals the fixed distance ξ_{ij} .

The moving frame, fixed point covariance function $\overline{u(y', t)u(y'', t-\tau)}$ can in principle then be obtained as the sum of pair contributions from all possible values of the fixed separation ξ_{ij} in the fluid. This leads to the summation over all values of (i) and (j):

$$\overline{\tilde{u}(y', t) \tilde{u}(y'', t-\tau)} = \sum_i^M \sum_j^M S(y', y'' | \xi_{ij}) \overline{\tilde{u}(y_i(t)) \tilde{u}(y_i(t-\tau) - \xi_{ij})} \quad (3.24)$$

In order to proceed, it is convenient to change the frame of reference for the two-particle covariance function on the right-hand side of Eq. (3.24) back to the fixed coordinate system, by use of the relation $\tilde{u}(y_i) = u(y_i + c) = u(x_i)$. The two-particle covariance in this way becomes

$$\overline{\tilde{u}(y_i(t)) \tilde{u}(y_i(t-\tau) - \xi_{ij})} = \overline{u(x_i(t)) u(x_i(t-\tau) - \xi_{ij})} \quad (3.25)$$

where the condition imposed on the two particles is that $x_i(t-\tau) = x_j(t-\tau) + \xi_{ij}$. As Fig. 5a shows, Eq. (3.25) expresses the correlation between the velocity of a fluid particle (i) at time t in the position x_i , and the velocity at time t- τ of the fluid particle (j) that is displaced by the distance ξ_{ij} relative to $x_i(t-\tau)$.

Alternatively, by referring the fixed distance ξ_{ij} separating the two particles to time t as shown in Fig. 5b, rather than to time t- τ , the covariance function between the two particles alternatively reads $\overline{u(x_j(t-\tau)) u(x_j(t) + \xi_{ij})}$, where now $x_i = x_j(t) + \xi_{ij}$. In the stationary and homogeneous turbulent field of consideration, these two alternative definitions must be identical, since the situation in Fig. 5b follows immediately from a time reversal of the situation in Fig. 5a. Moreover, these covariance functions will be independent of both the fluid particles absolute position x, as well as of the absolute time t. This leaves a function of the time lag τ and separation ξ_{ij} alone, which will be defined as

$$\begin{aligned} R_{\text{abs}}(\xi_{ij}, \tau) &= \overline{u(x_i(t)) u(x_i(t-\tau) - \xi_{ij})} \\ &= \overline{u(x_j(t-\tau)) u(x_j(t) + \xi_{ij})} \end{aligned} \quad (3.26)$$

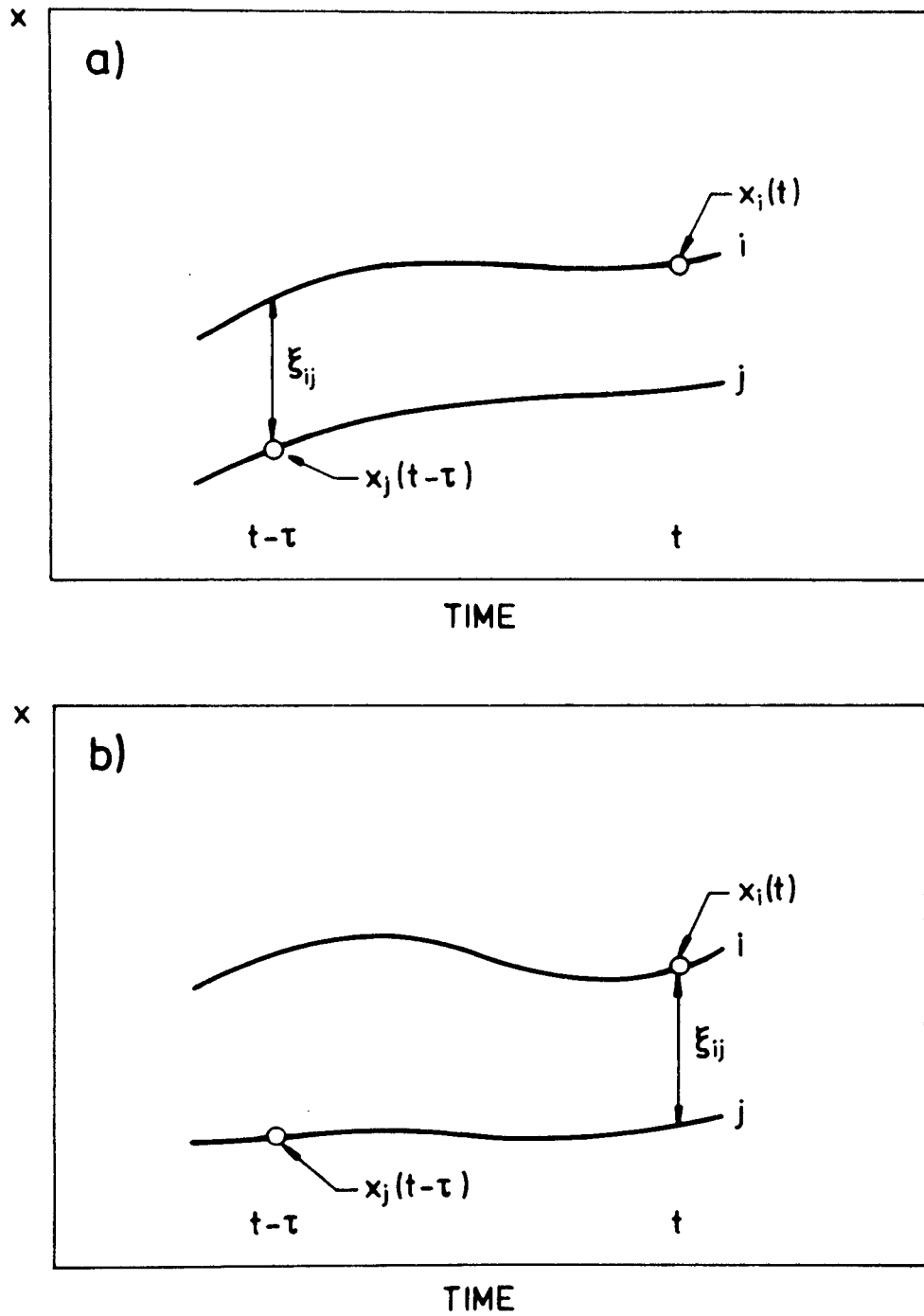


Fig. 5. The two-particle covariance function defined in Eq. (3.26). a) Referring the fixed particle separation ξ_{ij} to time $t-\tau$: $\overline{u(x_i(t))u(x_i(t-\tau)-\xi_{ij}(t-\tau))}$. b) Referring ξ_{ij} to time t : $\overline{u(x_j(t-\tau))u(x_j(t)+\xi_{ij}(t))}$. In homogeneous and stationary turbulence, these two definitions are identical.

Setting $\xi_{ij} = 0$ reduces this two-particle covariance to the Lagrangian auto-covariance function of a single particle: $R_{abs}(0, \tau) = R_{abs}(\tau)$, where $R_{abs}(\tau)$ was defined in Eq. (3.6). On the other hand, by setting $\tau = 0$, a pure Eulerian space-covariance results, for which the separation ξ_{ij} is along the direction of the velocity component u .

With both ξ_{ij} and τ set equal to zero, the two-particle covariance function yields the total energy $\overline{u^2}$ of the turbulence. $R_{abs}(\xi_{ij}, \tau)$ defines, with $\xi_{ij} = 0$, the Lagrangian integral time scale appropriate for a single particle through

$$t_L = (\overline{u^2})^{-1} \int_0^{\infty} R_{abs}(0, \tau) d\tau \quad (3.27)$$

Also, a (fixed point) Eulerian integral length scale for the turbulence can be obtained as

$$l_E = (\overline{u^2})^{-1} \int_0^{\infty} R_{abs}(\xi_{ij}, 0) d\xi_{ij} \quad (3.28)$$

The two-particle covariance function $R_{abs}(\xi_{ij}, \tau)$ somewhat resembles the two-particle Lagrangian covariance $\overline{u_1(t)u_2(\tau)}$ discussed in connection with Eq. (1.7). But where this covariance is restricted to two fluid particles, which at the time of release are located at the source position, the covariance in Eq. (3.26) has two arguments, namely the particle separation ξ_{ij} at a particular time and a time lag τ .

It remains to investigate for a particular pair of fluid particles, (i) and (j), the joint probability distribution $S(y', y'' | \xi_{ij})$ in Eq. (3.24) for finding the i'th fluid particle at y' at time t , and the j'th fluid particle at y'' at time $t - \tau$, under the condition that $y_i(t - \tau) = y_j(t - \tau) + \xi_{ij}$. We shall do so in terms of the previously discussed assumption about the underlying diffusion process, namely that the individual fluid particles in the moving frame follow identical and independent Gaussian statistics: Let d denote the (small) linear extent of a fluid particle and suppose that only one fluid particle at a

time can occupy the interval from $y-d/2$ to $y+d/2$ associated with a position y . At the time $t-\tau$, one particle out of the total of M will therefore hold the position associated with y'' , and the probability that this particle should happen to be the j 'th of the pair is simply $1/M$. If in this way $y_j(t-\tau) = y''$, then the i 'th particle of the pair holds the position $y_i(t-\tau) = y_j(t-\tau) + \xi_{ij}$ the fixed distance ξ_{ij} apart. The probability that this i 'th particle, in the time interval between $t-\tau$ and t , will reach inside the interval from $y'-d/2$ to $y'+d/2$ associated with the point y' is given by $G_S(y'-y_i(t-\tau), t) d$, where G_S is the Gaussian distribution of the relative displacement process Δy_i with standard deviation s as defined in Eq. (3.18) (see also Fig. 3).

For the particular particle pair considered, the following relations therefore apply

$$\begin{aligned} S(y', y'' | \xi_{ij}) &= 1/M G_S(y' - y_i(t-\tau), t) d \\ &= 1/M G_S(y' - (y_j(t-\tau) + \xi_{ij}), t) d \\ &= 1/M G_S(y' - y'' - \xi_{ij}, t) d \end{aligned} \quad (3.29)$$

By substituting this, together with the two-particle covariance function in Eq. (3.26), the following expression is obtained for the fixed point velocity covariance in Eq. (3.24):

$$\begin{aligned} \overline{u(y', t) u(y'', t-\tau)} &= \\ \sum_i^M \sum_j^M d/M G_S(y' - y'' - \xi_{ij}, t) R_{abs}(\xi_{ij}, \tau) \end{aligned} \quad (3.30)$$

If the number of fluid particles between $x_i(t-\tau)$ and $x_j(t-\tau)$ is denoted by $n = i-j$, the fixed particle pair separation can be written as $\xi_{ij} = nd$.

Further, it is possible to calculate the double sum in Eq. (3.30) as a sum over all possible values of i and j where the difference $n = i-j$ is fixed, followed by a sum over all n , viz.

$$\overline{\tilde{u}(y', t) \tilde{u}(y'', t - \tau)} =$$

$$\sum_{n=-M}^M \sum_{i=n+j} d/M G_S(y' - y'' - nd, t) R_{abs}(nd, \tau) \quad (3.31)$$

Both G_S and R_{abs} are decreasing functions of their argument nd . Therefore, by going to the limit for very large M , corresponding to an extension of the turbulent field to infinity on both sides of the diffusing cloud, only the terms for which $n \ll M$ will contribute to the double sum in Eq. (3.31). With n fixed at a value much smaller than M , the sum over $i = n+j$ approximately equals M times the argument in Eq. (3.31) and only the sum over the differences n remains

$$\overline{\tilde{u}(y', t) \tilde{u}(y'', t - \tau)} =$$

$$\sum_{n=-M}^M G_S(y' - y'' - nd, t) R_{abs}(nd, \tau) d \quad (3.32)$$

Finally, by letting the total number of particles M approach infinity at the same time as the extent d of the individual fluid particles becomes small (relative to the Kolmogorov scale of the turbulence), the pair separation $\xi_{ij} = nd$ can be considered a continuous independent variable ξ , and in its equivalent integral form, Eq. (3.32) therefore becomes

$$\overline{\tilde{u}(y', t) \tilde{u}(y'', t - \tau)} =$$

$$\int_{-\infty}^{\infty} G_S(y' - y'' - \xi, t) R_{abs}(\xi, \tau) d\xi \quad (3.33)$$

Here also d has been replaced by the differential increment $d\xi$. With this result it is now possible to calculate the centre-of-mass covariance function in Eq. (3.16). With the standard deviation of G_S as given in Eq. (3.18), the following integral has to be evaluated

$$R_{cm}(t, \tau) =$$

$$\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} R_{abs}(\xi, t) G_S(y' - y'' - \xi, t) G_{\sigma}(t)(y', t) G_{\sigma}(t - \tau)(y'', t - \tau) dy' dy'' d\xi \quad (3.34)$$

By keeping ξ fixed, the remaining two integrals are simply (double) convolutions of two Gaussian distribution functions. The result of this is another Gaussian with standard deviation equal to the square root of the sum of the individual variances: $\{(\sigma^2(t) - \sigma^2(t - \tau)) + \sigma^2(t) + \sigma^2(t - \tau)\}^{1/2} = \sqrt{2} \sigma(t)$. In this way, the final expression for the centre-of-mass covariance function becomes

$$R_{cm}(t, \tau) =$$

$$\int_{-\infty}^{\infty} R_{abs}(\xi, \tau) \frac{1}{2\sqrt{\pi} \sigma(t)} \exp \left\{ -\frac{1}{4} \frac{\xi^2}{\sigma^2(t)} \right\} d\xi \quad (3.35)$$

When an initially small puff is released, σ is much smaller than the Eulerian length scale ℓ_E and in this case $R_{cm}(t, \tau) \approx R_{abs}(0, \tau)$. This implies that the centre-of-mass covariance function, and consequently the centre-of-mass spread, equals that of a single particle in this limit.

In the other limit, when σ has grown to a size much greater than the length scale ℓ_E , $R_{cm}(t, \tau)$ becomes small compared with $R_{abs}(\tau)$. This implies that the centre-of-mass dispersion $\overline{c^2}$ becomes negligible in this far field limit, and that the relative diffusion (σ^2) is entirely dominated by single-particle diffusion ($\overline{x^2}$).

When the centre-of-mass covariance function Eq. (3.35) is inserted in Eq. (3.8), an implicit formula for the growth of a Gaussian puff results

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \int_0^t \left\{ R_{abs}(0, \tau) - \int_{-\infty}^{\infty} R_{abs}(\xi, \tau) \frac{1}{2\sqrt{\pi} \sigma(t)} \exp\left(-\frac{1}{4} \frac{\xi^2}{\sigma^2(t)}\right) d\xi \right\} d\tau \quad (3.36)$$

In the previous chapter (Eq. (2.24)), the cloud dispersion was expressed in terms of a mean-square relative velocity $\overline{v^2}(t)$ and a relative Lagrangian time scale $t_r(t)$, as

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \overline{v^2}(t) \cdot t_r(t) \quad (3.37)$$

From Eq. (3.35) with $\tau = 0$, and from Eq. (3.4), the mean-square relative velocity can now be identified as

$$\overline{v^2}(t) = R_{abs}(0, 0) - \int_{-\infty}^{\infty} R_{abs}(\xi, 0) \frac{1}{2\sqrt{\pi} \sigma(t)} \exp\left(-\frac{1}{4} \frac{\xi^2}{\sigma^2(t)}\right) d\xi \quad (3.38)$$

Equivalently, the relative correlation function $r(t, \tau)$ defined in Eq. (2.22) explicitly becomes

$$r(t, \tau) = \left(\overline{v^2}(t) \right)^{-1} \left\{ R_{abs}(0, \tau) - \int_{-\infty}^{\infty} R_{abs}(\xi, \tau) \frac{1}{2\sqrt{\pi} \sigma(t)} \exp\left(-\frac{1}{4} \frac{\xi^2}{\sigma^2(t)}\right) d\xi \right\} \quad (3.39)$$

With this correlation function given, the relative time scale $t_r(t)$ is easily obtained by an integration of $r(t, \tau)$ with respect to τ , as defined in Eq. (2.23).

3.2. Spectral formulation of relative diffusion

It is possible to introduce a spectral representation of the two-particle covariance function $R_{abs}(\xi, \tau)$ defined in Eq. (3.26).

The spectrum $S(k, \omega)$, where k is wavenumber and ω the frequency is defined by the Fourier transform

$$S(k, \omega) = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} R_{abs}(\xi, \tau) \exp(-i(k\xi + \omega\tau)) d\xi d\tau \quad (3.40)$$

In Appendix B it is shown that the single-particle Lagrangian spectrum $S_L(\omega)$, which is obtained by setting $\xi = 0$, is related to $S(k, \omega)$ through

$$\overline{u^2} S_L(\omega) = \int_{-\infty}^{\infty} S(k, \omega) dk \quad (3.41)$$

and also that the (fixed point) Eulerian spectrum $S_E(k)$, which results by setting $\tau = 0$, is related through

$$\overline{u^2} S_E(k) = \int_{-\infty}^{\infty} S(k, \omega) d\omega \quad (3.42)$$

The inverse Fourier transform corresponding to Eq. (3.40) is defined as

$$R_{abs}(\xi, \tau) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \exp(i(k\xi + \omega\tau)) dk d\omega, \quad (3.43)$$

It is can now be seen that the single-particle Lagrangian covariance function $R_{abs}(0, \tau)$ can be represented as

$$\begin{aligned} R_{abs}(0, \tau) &= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \exp(i\omega\tau) dk d\omega \\ &= \int_{-\infty}^{\infty} S_L(\omega) \exp(i\omega\tau) d\omega \end{aligned} \quad (3.44)$$

With these definitions, the growth rate of the cloud in Eq. (3.36) now becomes

$$\begin{aligned}
 \frac{1}{2} \frac{d\sigma^2}{dt} = & \int_0^t \left\{ \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \exp(i\omega\tau) d\omega dk \right. \\
 & - \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \exp(i(k\xi + \omega\tau)) \frac{1}{2\sqrt{\pi} \sigma(t)} \\
 & \times \exp\left(-\frac{1}{4} \frac{\xi^2}{\sigma^2(t)}\right) d\omega dk d\xi \Big\} d\tau
 \end{aligned} \tag{3.45}$$

The integration over ξ of the second term on the right-hand side is an inverse Fourier transform of the Gaussian distribution, i.e.

$$\begin{aligned}
 & \int_{-\infty}^{\infty} \frac{1}{2\sqrt{\pi} \sigma(t)} \exp\left(-\frac{1}{4} \frac{\xi^2}{\sigma^2(t)}\right) \exp(ik\xi) d\xi \\
 & = \exp\left(-k^2 \sigma^2(t)\right)
 \end{aligned} \tag{3.46}$$

By use of this in Eq. (3.45), the following equation results for the growth of a Gaussian cloud, expressed in terms of the spectrum $S(k, \omega)$ of the turbulence (see also Eq. (3.62)).

$$\begin{aligned}
 \frac{1}{2} \frac{d\sigma^2}{dt} = & \int_0^t \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \left(1 - \exp(-k^2 \sigma^2(t))\right) \\
 & \times \exp(i\omega\tau) d\omega dk d\tau
 \end{aligned} \tag{3.47}$$

By specifying $S(k, \omega)$, this equation can be solved for $d\sigma/dt$ as a function of time t , numerically at least, and also, upon a further integration over time from zero to t , for the cloud size $\sigma(t)$.

In Eqs. (3.36) and (3.45), the terms in the brackets $\{ \}$ equal the relative velocity covariance from Eq. (3.6) $\overline{v(t)v(t-\tau)}$. Analogous to the procedure used to arrive at Eq. (3.38), it is found by setting $\tau = 0$ that the mean-square relative velocity in the spectral representation can be expressed as

$$\begin{aligned}\overline{v^2(t)} &= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(\omega, k) \left(1 - \exp(-k^2 \sigma^2(t))\right) d\omega dk \\ &= \int_{-\infty}^{\infty} \overline{u^2} S_E(k) \left(1 - \exp(-k^2 \sigma^2(t))\right) dk\end{aligned}\quad (3.48)$$

This shows the important result that the mean-square relative velocity of the expanding cloud is closely related to the Eulerian space spectrum $S_E(k)$.

The relative correlation function, Eq. (3.39) correspondingly becomes, in terms of the spectrum $S(k, \omega)$

$$\begin{aligned}r(t, \tau) &= \\ &\left(\overline{v^2(t)}\right)^{-1} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(\omega, k) \left(1 - \exp(-k^2 \sigma^2(t))\right) \exp(i\omega\tau) d\omega dk\end{aligned}\quad (3.49)$$

The relative time scale $t_r(t)$ is, as before, obtainable from an integration with respect to τ , as defined in Eq. (2.23).

The equation for growth, Eq. (3.47), will next be considered in the limit where the cloud size σ is large compared to the length scale ℓ of the turbulence. Then, for all relevant values of k , the quantity $1 - \exp(-\sigma^2 k^2) \approx 1$ and by use of Eq. (3.41), there results in this limit

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \int_0^t \int_{-\infty}^{\infty} S_L(\omega) \exp(i\omega\tau) d\omega d\tau \quad (3.50)$$

Integrating twice with respect to time yields

$$\sigma^2(t) = t^2 \int_{-\infty}^{\infty} S_L(\omega) \frac{\sin^2(\frac{\omega t}{2})}{(\frac{\omega t}{2})^2} d\omega \quad (3.51)$$

This is simply G.I. Taylor's formula for single-particle diffusion. Not surprisingly, it is seen that the different behaviour

of the spread of a cloud, when compared with that of a single particle, is closely related to the spatial correlation of the turbulence.

In the limit where the time t is also large compared to the time scale t_L , Eq. (3.51) reduces to the usual far field limit $\sigma^2 = 2\overline{u^2}t_L t$, appropriate for single-particle dispersion.

3.3. Approximative solutions to the relative diffusion equation

Here will first be investigated the implications of an approximation similar to that suggested by G.I. Taylor (see Eq. (1.12)). Suppose that the two-particle covariance function in Eq. (3.26) can be replaced by a product of a fixed point Eulerian correlation function at time t : $\rho_E(\xi) = \overline{u(x,t)u(x+\xi,t)}/\overline{u^2}$ and a single-particle Lagrangian auto-correlation function $\rho_L(\tau) = \overline{u(x_i(t))u(x_i(t-\tau))}/\overline{u^2}$, in which case

$$R_{abs}(\xi, \tau) = \overline{u^2} \rho_E(\xi) \rho_L(\tau) \quad (3.52)$$

Even though Sawford (1982) found this type of approximation to be the most appropriate in his comparison, this approximation cannot in general be valid, and it is unlikely that it is particularly good except perhaps when τ is small compared to t_L . In this approximation the Fourier transform in Eq. (3.40) gives

$$S(k, \omega) = \overline{u^2} S_E(k) S_L(\omega) \quad (3.53)$$

where

$$S_E(k) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \rho_E(\xi) \exp(-ik\xi) d\xi \quad (3.54)$$

and

$$S_L(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \rho_L(\tau) \exp(-i\omega\tau) d\tau \quad (3.55)$$

When Eq. (3.53) is substituted into (3.47), the integral over ω is identified as the Lagrangian correlation coefficient $\rho_L(\tau)$ (see Appendix B, Eq. B5). We then write

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \int_0^t \rho_L(\tau) \left\{ \overline{u^2} \int_{-\infty}^{\infty} S_E(k) [1 - \exp(-k^2 \sigma^2(t))] dk \right\} d\tau \quad (3.56)$$

However, as before, the term in the brackets $\{ \}$ equals the mean-square relative velocity $\overline{v^2}(t)$ (cf. Eq. (3.48)). The remaining integral over τ is identified as the relative Lagrangian time scale $t_r(t)$ when comparison is made with Eq. (3.37).

Consequently, based on the approximation in Eq. (3.52), the following set of equations for the growth of a Gaussian cloud results:

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \overline{v^2}(t) \cdot t_r(t) \quad (3.57)$$

where

$$\overline{v^2}(t) = \overline{u^2} \int_{-\infty}^{\infty} S_E(k) [1 - \exp(-k^2 \sigma^2)] dk \quad (3.58)$$

and

$$t_r(t) = \int_0^t \rho_L(\tau) d\tau \quad (3.59)$$

A consequence of the "factorization" of $R_{abs}(\xi, \tau)$ into Eulerian and Lagrangian correlation functions is that the relative time scale becomes identical to the time scale appropriate for single-particle diffusion. The mean square relative velocity, however, is here, as well as under more general conditions (Eq. (3.48)), found to be related exclusively to the Eulerian properties of the turbulence.

One question that remains to be investigated is to what extent the estimate of the relative time scale in Eq. (3.59) applies to common turbulence.

Starting with the limit for large times where $t \gg t_L$, the relative time scale $t_r(t)$ in Eq. (3.59) becomes equal to t_L as it properly should, when the particles move independently of each other. In the small time limit, on the other hand, the approximate solution to Eq. (3.59) yields

$$t_r(t) = t \quad \text{for} \quad t \ll t_L \quad (3.60)$$

since $\rho_L(\tau) \approx 1$ for small time lags. The extent that this limiting value is consistent with the more general solution, Eq. (3.47) can be examined, viz.

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \int_0^t \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \left(1 - \exp(-k^2 \sigma^2(t))\right) \exp(i\omega\tau) d\omega dk d\tau \quad (3.61)$$

Without loss of generality, an integration over the time lag τ reduces this equation to the simpler form

$$\frac{1}{2} \frac{d\sigma^2}{dt} = t \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \frac{\sin(\omega t)}{\omega t} \left(1 - \exp(-k^2 \sigma^2(t))\right) d\omega dk \quad (3.62)$$

When the time t is sufficiently small, the sinc function $\sin(\omega t)/\omega t$ remains close to unity for all values of the angular frequency ω , where $S(k, \omega)$ contributes to the integral (see Fig. 6). Therefore, the following approximation must apply applicable in the small time limit:

$$\int_{-\infty}^{\infty} S(k, \omega) \frac{\sin(\omega t)}{\omega t} d\omega \approx \int_{-\infty}^{\infty} S(k, \omega) d\omega = \overline{u^2} S_E(k) \quad (3.63)$$

With this approximation, Eq. (3.62) in the limit of $t \ll t_L$ becomes

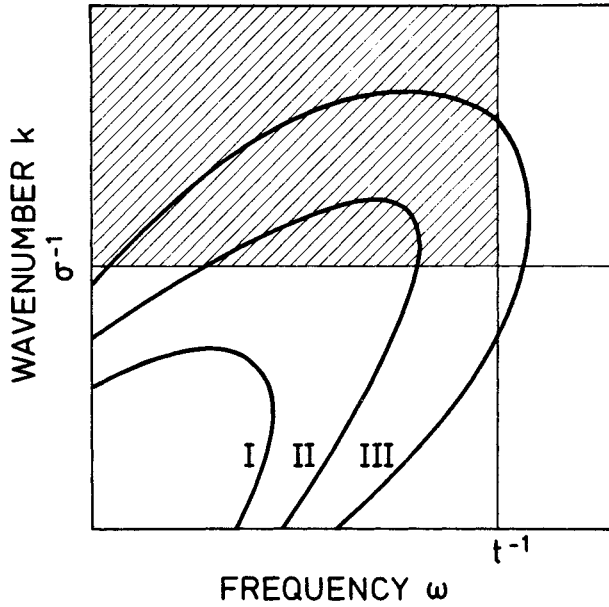


Fig. 6. Iso-contour plot of a hypothetical spectrum $S(k, \omega)$. Its maximum value is at $(k, \omega) = (0, 0)$ from where the function monotonically decreases through the levels I, II, and III. The cutoff frequency associated with the low-pass filter $\sin(\omega t)/\omega t$ is schematically drawn as the vertical line at $\omega = t^{-1}$. Correspondingly, the high-pass filter $(1 - \exp(-k^2 \sigma^2))$ essentially cuts away wavenumbers that are smaller than σ^{-1} . The shaded area therefore represents the part of the spectrum $S(k, \omega)$ that essentially contributes to the integral over k and ω in Eq. (3.44).

$$\begin{aligned} \frac{1}{2} \frac{d\sigma^2}{dt} &= t \overline{u^2} \int_{-\infty}^{\infty} S_E(k) \left(1 - \exp(-k^2 \sigma^2(t)) \right) dk \\ &= t \cdot \overline{v^2}(t) \end{aligned} \quad (3.64)$$

It is also seen that the small limiting value for $t_r(t)$ from Eq. (3.60) is consistent with the general solution in Eq. (3.47).

For values of t in the interval between the near and far field limits, the degree of approximation associated with $t_r(t)$ when estimated from Eq. (3.59), depends on the statistical dependence between the two variables ω and k . If ω and k are totally independent of each other, then $S(k, \omega) = \overline{u^2} S_E(k) S_L(\omega)$, and consequently the quantity (Tennekes and Lumley (1972) p. 207) $\overline{\omega k} = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \omega k S(k, \omega) d\omega dk / \overline{u^2}$ is equal to zero. On the other hand, finding $\overline{\omega k} = 0$ in a particular turbulent field is only a necessary, but not a sufficient condition for independence and for the applicability of Eq. (3.59).

There are situations, however, where it is unnecessary to be concerned about the general applicability of Eq. (3.59). This is when the cloud growth is dominated entirely by the Eulerian

properties of the turbulence, which is the case when the function $\overline{v^2}(t)$ plays an all dominant role for the dispersion on a relatively short period of time after the release at $t = 0$. In that case $t_r(r) \approx t$ ($\ll t_L$) is a reasonably good approximation for the relative time scale, and the growth of the cloud can be calculated simply on the basis of Eq. (3.64).

3.4. Spreading of Gaussian puffs related to Eulerian power law spectra

When considering diffusion times that are small relative to the Lagrangian integral time scale of the turbulence t_L , it was shown above that the growth of the Gaussian puff is determined by the simple set of equations

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \overline{v^2(t)} \cdot t_r(t) \quad (3.65)$$

$$\overline{v^2(t)} = \overline{u^2} \int_{-\infty}^{\infty} S_E(k) \left(1 - \exp(-k^2 \sigma^2(t)) \right) dk$$

$$t_r(t) = t \quad \text{for} \quad t \ll t_L$$

In the limit for small times, only the Eulerian properties of the turbulence (through the wavenumber spectrum $S_E(k)$), therefore, come into play. The set of equations (3.65) will now be investigated analytically by assuming that the Eulerian wavenumber spectrum is given as a power law $S_E(k) = \delta k^p$, where δ is a constant of dimension $m^{(1+p)}$. Spectra characterized by $p \geq -1$ results in divergence of the otherwise normalized integral $\int_{-\infty}^{\infty} S(k) dk = 1$. Such powers can consequently be included in the analysis only as subranges of limited extension. A power law representation of the Eulerian wavenumber spectrum $S_E(k)$ is also of relevance over only limited ranges of wavenumbers. For instance, at very small wavenumbers ($k \sim 0$), the theoretical spectrum tends to be flat ($p = 0$), and approaches the amplitude level $\sim \ell_E/\pi$ asymptotically.

For cases where the power p lies within the interval: $-3 < p < -1$, the second of the set of equations (3.65) can be integrated by parts to give

$$\overline{v^2}(t) = - \frac{2\overline{u^2}\delta}{(p+1)} \Gamma\left(\frac{p+3}{2}\right) \sigma^{-(p+1)}, \quad \text{for } -3 < p < -1 \quad (3.66)$$

where Γ denotes the gamma function.

For cases where $p \leq -3$, the integral in Eq. (3.65) for determining $\overline{v^2}$ is divergent. This occurs because the integration with $S_E(k) = \delta k^p$ extends unphysically over all k and not only over a limited subrange as discussed above. In order to remedy this problem an approximation of the high-pass filter $(1 - \exp(-\frac{1}{2} k^2 \sigma^2(t)))$ by Heaviside's step function, has been introduced*

$$H(k) = \begin{cases} 0 & \text{for } |k| < 1/\sigma \\ 1 & \text{for } |k| \geq 1/\sigma \end{cases} \quad (3.67)$$

The relative velocity variance in this case simply becomes

$$\overline{v^2}(t) = - \frac{2\overline{u^2}\delta}{(p+1)} \sigma^{-(p+1)}, \quad \text{for } p \leq -3. \quad (3.68)$$

The differential equation for $\sigma(t)$ in Eq. (3.65) is now readily solved.

The following basically different solutions are found, all of which are applicable only in the limit $t \ll t_L$.

*This corresponds to a "top hat" rather than a Gaussian cloud.

i) for $-3 < p < -1$

$$\sigma(t) = \{ct^2 + \sigma_0^{1/q}\}q$$

ii) for $p = -3$

$$\sigma(t) = \sigma_0 \exp\left(\frac{1}{2} \overline{\delta u^2} t^2\right) \quad (3.69)$$

iii) for $p < -3$

$$\sigma(t) = \{\tilde{c} t^2 + \sigma_0^{1/q}\}q$$

Here, $q = 1/(3+p)$, $\tilde{c} = -\overline{u^2} \delta(3+p)/(1+p)$ and $c = \tilde{c} \Gamma((p+3)/2)$. σ_0 is the initial size of the cloud, i.e. $\sigma(t=0)$. In order that solution iii) for $p < -3$ apply, it must be required that $t < t_{\max}$, where $t_{\max} = (\sigma_0^{1/q}/|\tilde{c}|)^{1/2}$. The limit $t = t_{\max}$, however, will never be approached with a physically meaningful energy spectrum specified at the smallest wavenumbers.

The behaviour of $\sigma(t)$ in the phase of spread, where the initial puff size σ_0 is an important parameter, can also be deduced from Eq. (3.65) by substituting Eqs. (3.66) for $\overline{v^2}(0)$ with $\sigma = \sigma_0$. For $t \ll \{\sigma_0^2/\overline{v^2}(0)\}^{1/2}$, a second-order expansion of the initial dispersion reads

$$\sigma^2(t) = \sigma_0^2 + \overline{v^2}(0) \cdot t^2 \quad (3.70)$$

In form this equation is similar to Eq. (1.10), and is thus in accordance with the result of Batchelor's similarity theorem in the near-field limit.

Within the time interval described by Batchelor as "intermediate", i.e. when viscosity and the initial puff size are no longer dominant, but before the integral time scale t_L becomes an important scaling parameter, the first of Eqs. (3.69) yields

$$\sigma(t) = c q t^{2/(3+p)} \quad (3.71)$$

where the constraints are: $-3 < p < -1$ and $\{\sigma_0^2/\overline{v^2}(0)\}^{1/2} \ll t \ll t_L$.

In the following chapter the implications to atmospheric dispersion of the set of Eqs. (3.65) and their solutions Eq. (3.69) will be discussed.

4. APPLICATION TO ATMOSPHERIC DISPERSION

4.1. Relative diffusion within the inertial subrange

Turbulence in the inertial subrange of the atmospheric boundary layer is often represented in terms of Eulerian wavenumber spectra in the non-normalized form

$$\overline{u^2} S_E(k) = \alpha \epsilon^{2/3} k^{-5/3} \quad (4.1)$$

Here α is a constant of order unity and ϵ being the rate of dissipation of energy. Setting $\delta = \alpha \epsilon^{2/3}/\overline{u^2}$ and $p = -5/3$, Eq. (3.71) for the growth of a cloud becomes

$$\sigma^2(t) = (2 \Gamma(\frac{2}{3}) \alpha)^{3/2} \epsilon t^3 \quad (4.2)$$

applicable for "intermediate" times only as defined in Eq. (3.71). When compared with Eq. (1.11), this result is also found to be in agreement with Batchelor's inertial subrange theory on relative diffusion.

For the case of homogeneous and isotropic turbulence, Tennekes and Lumley (1972) suggest the value of the constant $\alpha = \frac{9}{55} \times 1.5 = 0.246$ for the wavenumber spectrum $S_E(k)$ in question. (It should be emphasized that the proper one-dimensional spectrum to be used here is the so-called longitudinal spectrum, and not the corresponding transverse spectrum (see Tennekes and Lumley (1972)

p. 251 for precise definitions). This is because the velocity u is parallel to the particle separation ξ in Eq. (3.26).

For inertial subrange isotropic and homogeneous turbulence, the prediction for the spread of a Gaussian puff therefore becomes

$$\begin{aligned}\sigma^2(t) &= (1.34 \times 2 \times \frac{9}{55} \times 1.5)^{3/2} \epsilon t^3 \\ &= 0.534 \frac{\overline{u^2}}{t_L} t^3\end{aligned}\tag{4.3}$$

where the dissipation rate ϵ for later comparison has been replaced by $\overline{u^2}/t_L$.

Independently, F.B. Smith (1968) and F. Gifford (1981) have derived corresponding formulas for the instantaneous spread of a plume in short periods $t \ll t_L$

$$\sigma^2 = \frac{2}{3} \epsilon t^3\tag{4.4}$$

Their numerical coefficient is slightly larger than the one found in Eq. (4.3). Their models, however, describe the spread of individually released particles, the velocity of which in the fixed frame is governed by a Langevin equation with a specified initial velocity, common to all the particles released. Their model result (Eq. 4.4) thus describes the ensemble averaged spread of conditionally released single-particle diffusion rather than a real two-particle or relative diffusion process. Further, their model result is a consequence of an assumed Lagrangian exponential correlation function, the Fourier transform of which, when expressed in Eulerian terms, becomes

$$\overline{u^2} S_E(k) = \frac{\overline{u^2}}{\pi} \frac{l_E}{1 + (l_E k)^2}\tag{4.5}$$

This spectrum is representative for a $k^{-5/3}$ law only in a rather limited wavenumber interval in the neighbourhood of $(k l_E)^2 = 5$. In this case, Eq. (4.5) can be approximated by

$$u^2 S_E(k) = \frac{5^{5/6}}{6\pi} \overline{u^2} \epsilon_E^{-2/3} k^{-5/3} \quad (4.6)$$

Setting $\alpha = 5^{5/6}/6\pi = 0.203$, the relative diffusion model, Eq. (4.3) derived here yields a result which compares with an exponential correlation function

$$\sigma^2 = 0.401 \epsilon t^3 \quad (4.7)$$

In this case also, a notably smaller coefficient is found compared to the conditional single-particle result of Eq. (4.4).

When two particles are simultaneously released from a source with negligible (but non-zero) initial separation, both of them are immersed into one and the same coherent eddy structure. Their motion will thus remain coherent over a longer period of time than will be the case with singly released particles, immersed into individual eddy structures and correlated through a common initial velocity only. Being more correlated, the two simultaneously released particles will not diffuse as rapidly as the independently released particles. This constitutes a possible explanation for the somewhat different coefficients found in Eqs. (4.4) and (4.7).

4.2. Relative diffusion within the enstrophy inertial subrange

Two-dimensional turbulence theory has attracted wide-spread interest among meteorologists following the work of Kraichnan (1967) and others. The theoretical studies by Kraichnan of two-dimensional turbulence have shown that a source of energy and enstrophy (half-squared vorticity) isolated at wavenumber k_i leads to a wavenumber spectrum with a discontinuity at k_i . For $k < k_i$ energy is cascaded to lower wavenumbers and $S_E(k) \propto \epsilon^{2/3} k^{-5/3}$ and for $k > k_i$, enstrophy is cascaded to larger wavenumbers and $S_E(k) \propto \eta^{2/3} k^{-3}$, where η is the enstrophy cascade rate. In the latter range, the characteristic time scale T_c is $\eta^{-1/3}$. In contrast to eddy time scales in three-dimensional turbulence, this two-dimensional time scale, characteristic for

the small eddies in two-dimensional flow, is independent of the scale of motion. In the atmosphere, T_c is typically ~ 1 day. Several authors have provided evidence for the existence of the k^{-3} law in large-scale atmospheric spectra down to scales ~ 100 km (see, for instance, K.S. Gage (1979) for a recent summary).

By dimensional analysis, J.T. Lin (1972) obtained an exponential power law for relative diffusion in the enstrophy cascade range, by postulating that the relative diffusivity depends on both the local mean-square relative distance $\overline{\ell^2}$ and the enstrophy cascade rate η . By dimensional analysis

$$\frac{1}{2} \frac{d\overline{\ell^2}}{dt} = \gamma \eta^{1/3} \overline{\ell^2} \quad (4.8)$$

$$\overline{\ell^2} = \ell_0^2 \exp(2t/T_c) , \quad \text{for } t \gg T_c$$

where γ is a dimensionless coefficient of order unity and ℓ_0^2 the initial separation of two diffusing particles. Since T_c is considered a relevant time scale in Lin's dimensional analysis, it is implicitly assumed that $t \gg T_c$ in Eq. (4.8).

On the other hand, in the limit where the diffusion time t is small relative to T_c , t itself must be a proper scaling parameter for the relative time scale, i.e. $t_r \propto t$ (as also can be seen from Eq. (2.23)). The mean-square relative velocity $\overline{v^2}$ scales then with the mean-square separation and the fixed time T_c , so the larger the separation, the larger also is the relative variance, $\overline{v^2}$. Based upon the relative diffusivity $\overline{v^2} \cdot t_r$, dimensional analysis now gives

$$\frac{1}{2} \frac{d\overline{\ell^2}}{dt} = \gamma \frac{\overline{\ell^2}}{T_c^2} \cdot t \quad (4.9)$$

$$\overline{\ell^2} = \ell_0^2 \exp(t^2/T_c^2) , \quad \text{for } t \ll T_c$$

In the more familiar case of single-particle diffusion, characterized by an integral time scale t_L and a constant variance $\overline{u^2}$, dimensional analysis also yields two basically different solutions for the spread $\overline{x^2}$, analogous to Eqs. (4.8) and (4.9). When $t \gg t_L$, the rate of growth $1/2 \, d\overline{x^2}/dt$ is proportional to the (absolute) diffusivity $\overline{u^2} \cdot t_L$ whereas, when $t \ll t_L$, it is proportional to $\overline{u^2} \cdot t$. This gives rise to the two well-known sub-ranges for the spread of a single particle: $\overline{x^2} \propto t$ and $\overline{x^2} \propto t^2$, respectively.

On comparing the solution Eq. (3.69) for $p = -3$ with the dimensional analysis Eq. (4.9), it is found that the two solutions are consistent in that they have identical forms and that both of them applies to times that are small relative to the time scale of the turbulence.

In order to be able to compare the turbulent diffusion model suggested here with the dimensional result, Eq. (4.8), the time scale $t_r(t)$ in Eq. (3.65) is now set equal to T_C corresponding to the limit where $t \gg T_C$. Integrating the first of the equations (3.65) with $\overline{v^2}(t)$ as given by Eq. (3.68) for $p = -3$ results in

$$\sigma(t) = \sigma_0 \exp(t/T_C) \quad \text{for} \quad t \gg T_C \quad (4.10)$$

This is consistent with the result of J.T. Lin's (1972) dimensional analysis for relative diffusion in the enstrophy cascade subrange. Equation (4.10) applies to situations where the diffusion time t is large compared to the turbulent time scale T_C . At the same time, the puff size σ must be small compared to the length scale ℓ_E of the turbulence.

Figure 7 shows a summary of the four different regimes of diffusion predicted with a k^{-3} power law. The spectrum is assumed to be constant for $k < 1/\ell_E$. Note that the asymptotical values of $\sigma(t)$ equal that of single-particle diffusion, when $\sigma > \ell_E$. This example emphasizes the importance of distinguishing between length and time scales when dealing with relative diffusion.

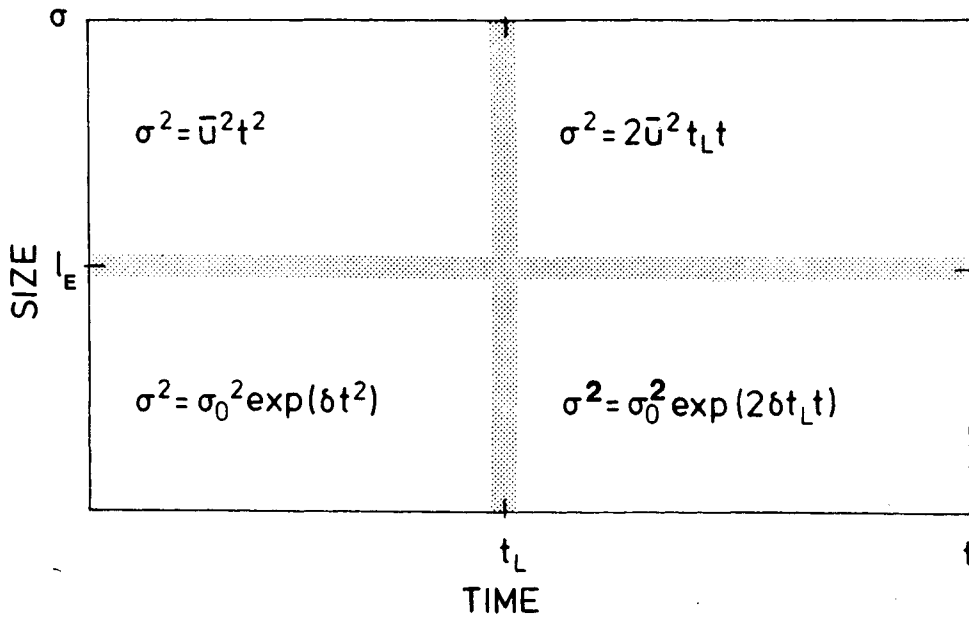


Fig. 7. The four regimes divided by the length scale l and time scale t_L in a space-time plot of relative diffusion, based on a power-law spectrum $S_E(k)$ that is proportional to k^{-3} for $k > 1/l_E$ and of constant amplitude for $k < 1/l_E$.

4.3. Relative diffusion within the troposphere

In Fig. 8 a schematic one-dimensional wavenumber spectrum has been composed from the literature, showing the different sub-ranges previously discussed. The diffusion of an initially small Gaussian cloud starts in the 3-dimensional isotropic inertial subrange and grows from there into the reverse energy cascading $k^{-5/3}$ inertial range of two-dimensional turbulence. By associating a sink rather than a source for enstrophy and energy at the 1000-km scale shown, the empirical data composed in Fig. 8 becomes consistent with the theory of Kraichnan (1967) previously discussed. After reaching a size $\sigma \sim 10^6$ m, the cloud grows into the k^{-3} enstrophy cascade subrange and ultimately, on the 10^7 m scale, the spectrum is assumed to level off.

By choosing the mean small-scale energy dissipation rate as low as $1-2 \cdot 10^{-4} \text{ m}^2 \text{ s}^{-3}$, the spectrum becomes almost a straight line at the interface between two- and three-dimensional turbulence. This occurs because the universal constant for the two-dimen-

sional upscale transport spectrum α_{II} is much larger than α_I belonging to the three-dimensional decay spectrum. Values for the longitudinal wavenumber spectrum shown are chosen as: $\alpha_I = 0.25$, $\alpha_{II} = 2.2$ and $\epsilon = 0.8 \cdot 10^{-3} \text{ m}^2 \text{ s}^{-3}$. At $k = 2\pi/1000 \text{ m}^{-1}$, $\overline{u^2 S_E}(k) = 10 \text{ m}^3 \text{ s}^{-2}$ and the rate of energy injection at $\lambda \sim 1000 \text{ m}$ is $\overline{u^2} dS_E/dt = 3.1 \cdot 10^{-5} \text{ m}^2 \text{ s}^{-3}$. With the sink at $\lambda = 10^6 \text{ m}$, the time scale $T_c = \eta^{-1/3}$ can also be determined to be of the order ~ 17 hours. The energy of the spectrum in the $-5/3$, the -3 and the flat part is $3\pi \text{ m}^2 \text{ s}^{-2}$, $99\pi \text{ m}^2 \text{ s}^{-2}$, and $200\pi \text{ m}^2 \text{ s}^{-2}$, respectively, and the corresponding length scale $\pi S(0) \approx 3140 \text{ km}$.

By calculating $\sigma(t)$ in the $-5/3$ subranges on the basis of Eq. (4.3), the cloud's travel time t will exceed the enstrophy integral time scale T_c at the $\sim 10^5 \text{ m}$ scale. Equation (4.10), applicable for $t \gg T_c$, is hence the appropriate formula, rather than Eq. (4.9), for a determination of the asymptotical form of $\sigma(t)$ in the -3 enstrophy cascade subrange.

Based on Eqs. (3.57)-(3.59), and on the spectrum in Fig. 8, not only the asymptotical form, but also the inter-regional growth of the Gaussian cloud can be determined on the basis of the spectrum in Fig. 8. However, the Eulerian wavenumber spectrum gives no information either about the Lagrangian correlation coefficient $\rho_L(\tau)$ or, as a consequence, about the relative time scale t_r in Eq. (3.59). For this reason, the following simple interpolation model for t_r is proposed for use here

$$t_r(t) = \frac{t_L}{1+t_L/t} \quad (4.11)$$

Here the quantity t_L is the parameter value of the integral time scale appropriate for large-scale dispersion (Gifford, 1982). The suggested function has the appropriate asymptotical forms, i.e. $t_r \approx t$ for $t \ll t_L$ and $t_r = t_L$ for $t \gg t_L$ as discussed previously in connection with Eqs. (3.59) and (3.60). The condition $t_r \leq t_L$ is fulfilled by Eq. (4.11). In addition, as long as $t_r(t)$ is chosen to be a smooth and monotonically increasing function of time, its specific form influences only the growth marginally, as is well known from G.I. Taylor's (1921) single-particle diffusion theory.

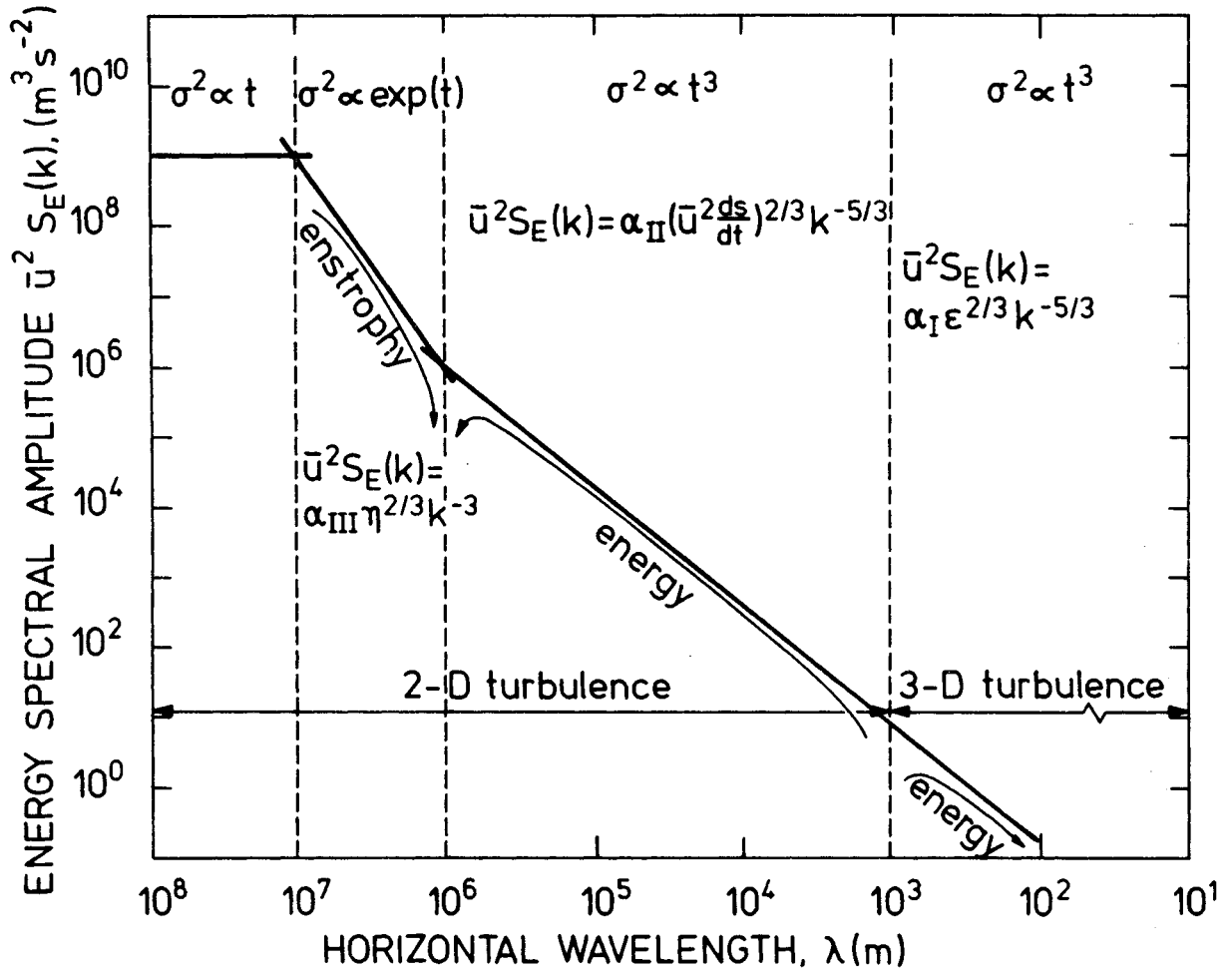


Fig. 8. Summation of relative diffusion $\sigma(t)$ of an initially small Gaussian cloud in relation to a schematic wavenumber spectrum composed from the literature by K.S. Gage, 1979, D.K. Lilly and E.L. Petersen, 1983, and D.K. Lilly, 1983. (See text (Chapter 4.3) for description.)

By use of the following set of substitutions

$$\tilde{\sigma}^2 = \sigma^2 / \Delta^2 \quad \text{where} \quad \Delta^2 = \overline{u^2} t_L^2 \quad (4.12)$$

$$\tilde{t} = t / t_L ; \quad \tilde{t}_r = t_r / t_L = \tilde{t} / (\tilde{t} + 1) ,$$

Equation (3.57) can now be written in the following dimensionless form, appropriate for numerical integration

$$\frac{1}{2} \frac{d\tilde{\sigma}^2}{d\tilde{t}} = \tilde{t}_r(\tilde{t}) \int_{-\infty}^{\infty} S_E(k) (1 - \exp(-k^2 \tilde{\sigma}^2 \Delta^2)) dk \quad (4.13)$$

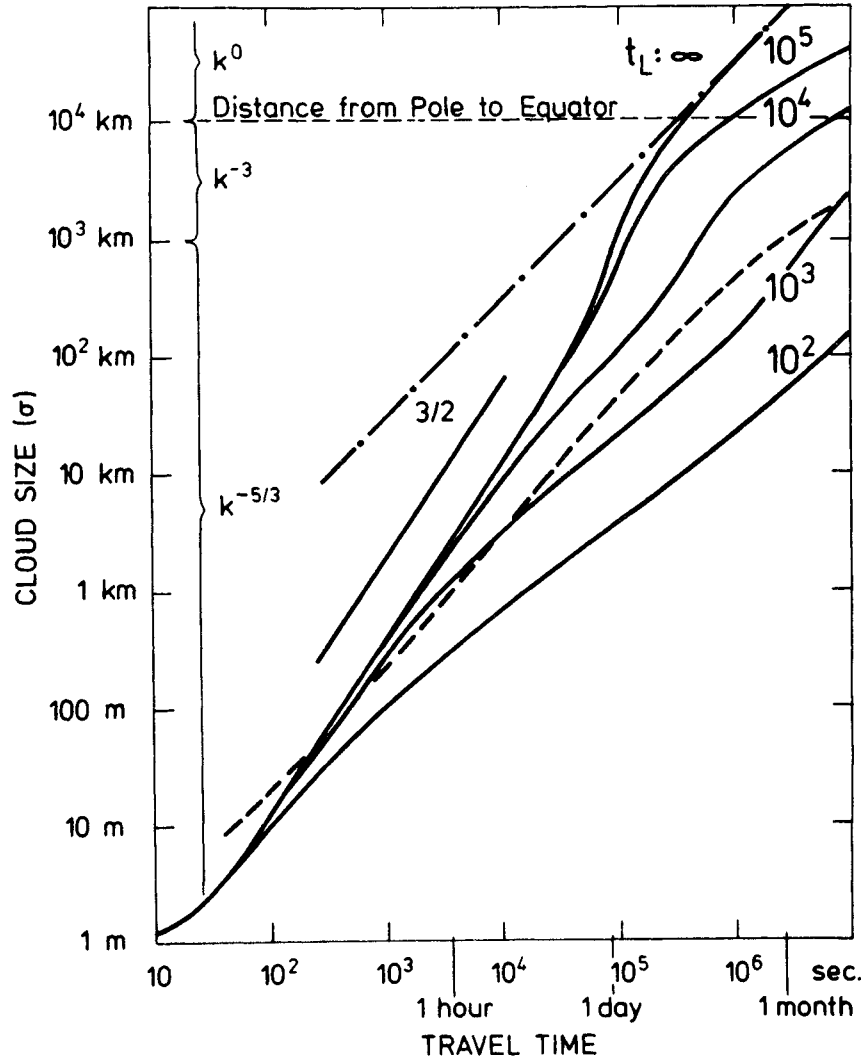


Fig. 9. Plot of cloud size σ for various values of the Lagrangian integral time scale t_L vs. travel time t , according to Eq. (4.12) and the energy spectrum of Fig. 8. The dashed curve (see Hage et al. (1967)) illustrates an empirical curve of horizontal atmospheric diffusion data over the entire atmospheric range. The maximum single-particle diffusion coefficient, $(\overline{u^2})^{1/2}t$, corresponding to the case where $t_L = \infty$, is shown as the topmost dashed-dotted line (see also Mikkelsen and Troen (1981)).

A single "universal" curve for $\bar{\sigma}(\tilde{t})$ is unobtainable from this nonlinear integro-differential equation. However, solutions can be found as a function of the single parameter $\Delta^2 = \overline{u^2}t_L^2$.

In Fig. 9 solutions to Eq. (4.13) are shown, where $\overline{u^2}$ and $S_E(k)$ corresponds to Fig. 8 and for various values of the integral

scale t_L . For all cases shown, the initial puff size $\sigma(0)$ was taken to be 1 metre.

For travel times t that are smaller than, say, 30 s, the numerical solution of $\sigma(t)$ follows the near-field limit of Eq. (3.69i) and for "intermediate" times when $t_L > 10^5$ s, the spread $\sigma(t)$ continues to follow the prediction in Eq. (4.2) ($\sigma = 0.0123 t^{3/2}$) up to $t \approx 10^4$ s. Then the cloud enters the exponential growth regime and the far field limit ($\sigma^2 \propto t$) is ultimately reached when $t \gg t_L$ and $\sigma > 10^4$ km.

Values of t_L smaller than $\sim 10^5$ s significantly alter the general behaviour of the growth with time as shown. For t_L less than approximately 100 s, the "intermediate" $3/2$ -region does not exist. In the literature, values of t_L range from 500 to $2 \cdot 10^5$ s (Gifford, 1982). A simple, but very crude estimate based on Pasquill's β -method is: $t_L \approx \beta \ell / (\overline{u^2})^{1/2}$. Taking $\beta = 4$ and ℓ and $\overline{u^2}$ from Fig. 8, $t_L = 4 \cdot 3140 \cdot 10^3 / 30 = 4 \cdot 10^5$ s. When a comparison is made with the empirical curve in Fig. 9 of horizontal atmospheric diffusion data, taken from Hage et al. (1967), this value of t_L seems rather high. A time scale of the order ~ 1 hour (3600 s) fits the empirical data better. At small wavenumbers, the spectral values in Fig. 8, and therefore also the energy $\overline{u^2}$ and the length scale ℓ of the hypothetical spectrum, are possibly unrealistically high, and smaller values of S_E in this domain might result in better agreement with the empirical curve, when t_L is calculated by the Pasquill β -method.

Before any final conclusion on the relative diffusion theory is drawn from the study here, however, it should be emphasized that the model Eq. (4.13) used for the computation of $\sigma(t)$ in Fig. 9 is a rather simplified version of the more general theory (Eq. (3.36)). In summary, the simplifications involved here are that the two-particle covariance function $R_{abs}(\xi, t)$ has been written as a product $\overline{u^2} \rho_L(\tau) \rho_E(\xi)$. The integral, Eq. (3.59), of $\rho_L(\tau)$ has then been modelled by Eq. (4.11), whereas $\rho_E(\xi)$ is specified through the inverse Fourier transform of $S_E(k)$ in Fig. 8.

Finally, Fig. 9 shows the single-particle diffusion coefficient, $(\overline{u^2})^{1/2} \cdot t$, corresponding to the case where $t_L = \infty$ and an infinite averaging time. As discussed, for instance, by Mikkelsen and Troen (1981), this coefficient represents an upper limit for σ in the far field limit. The condition that the single-particle diffusion coefficient represents an upper limit for σ is seen to be fulfilled in Fig. 9, whereas the corresponding value in the relative diffusion study by Sheih (1980) was exceeded by a factor of 3.

5. DISCUSSION

Derivation of analytical solutions for the turbulent dispersion of a cloud, in terms of the two-particle covariance function (Eq. (3.26)), or in terms of its corresponding spectrum (Eq. (3.47)), was made possible by assuming a non-fluctuating Gaussian particle distribution function. Inclusion of non-zero concentration fluctuations C' in the analysis, so that $C = \overline{C} + C'$ and $\overline{C'^2} > 0$ would inevitably have introduced terms in the analysis of the form (in Eq. (3.11) and onward)

$$\overline{u(x_i, t)u(x_j, t-\tau)C'(x_i, t)C'(x_j, t-\tau)} \quad (5.1)$$

together with third-order covariances of the variates u and c' as well. Therefore, it is expected that the number as well as quality of the assumptions required by conventionally modelling such terms (using eddy diffusivities) probably would have introduced at least as much uncertainty, if not even more, as is introduced here by setting $C' = 0$.

There does not seem to exist much reported observation of the mean-square of the fluctuations in concentration $\overline{C'^2}$ in clouds, but experimental evidence for steady plumes summarized by Csanady (1973) suggests that the distribution of $\overline{C'^2}$ is self-

similar and that the ratio to the square of the mean concentration $\overline{C'^2}/\overline{C}^2$ has a value at the centre which varies significantly from experiment to experiment (but typically somewhat less than 0.5) and then increases outwards, reaching values of order ~ 10 at the outer edge of the instantaneous plume.

Chatwin and Sullivan (1979) considered the mean square of the fluctuation in concentration $\overline{C'^2}$ and the ratio $\overline{C'^2}/\overline{C}^2$. The main theme of their paper is the way in which \overline{C} , $\overline{C'^2}$, and $\overline{C'^2}/\overline{C}^2$ vary in space and time. In terms of the fluid velocity vector \underline{v} relative to the moving origin \underline{c} of a cloud, the Eulerian mass balance over a stationary volume elements reads

$$\frac{\partial C}{\partial t} + \nabla \cdot (C \underline{v}) = \kappa \nabla^2 C \quad (5.2)$$

Here $\nabla \cdot$ and ∇^2 are the divergence and the Laplacian operators in the moving frame, respectively, and κ the molecular diffusivity. The instantaneous concentration of the cloud can be written in terms of its ensemble means and fluctuations as follows:

$$C(y, t) = \overline{C}(y, t) + C'(y, t), \quad \overline{C'} = 0 \quad (5.3)$$

For the relative velocity in the moving frame, we conclude from Eq. (2.19) that $\underline{v} = 0$, so

$$\underline{v} = \underline{v}'(y, t), \quad \overline{\underline{v}'} = 0 \quad (5.4)$$

Substitution of Eqs. (5.3) and (5.4) into (5.2) leads in the usual way (Reynolds decomposition) to the following equations

for \overline{C} and C' :

$$\frac{\partial \overline{C}}{\partial t} + \nabla \cdot (\overline{\underline{v}' C'}) = \kappa \nabla^2 \overline{C} \quad (5.5)$$

and

$$\frac{\partial C'}{\partial t} + \nabla \cdot (\underline{v}' \bar{C} + \underline{v}' C' - \overline{\underline{v}' C'}) = \kappa \nabla^2 C' \quad (5.6)$$

The equation for $\overline{C'^2}$ is obtained from Eq. (5.6) by multiplying by $2C'$, assuming incompressibility ($\nabla \cdot \underline{v} = 0$) and taking the ensemble mean. After rearranging, it becomes

$$\begin{aligned} \frac{\partial \overline{C'^2}}{\partial t} = & - 2 \overline{\underline{v}' C'} \nabla \bar{C} \\ & + \nabla \cdot (\kappa \nabla \overline{C'^2} - \overline{\underline{v}' C'^2}) \\ & - 2 \kappa \overline{(\nabla C')^2} \end{aligned} \quad (5.7)$$

The first term on the right-hand side is conventionally described as the production of $\overline{C'^2}$ (by feeding from the distribution of \bar{C} through the mechanism described by the term in Eq. (5.5) involving $\underline{v}' C'$). The divergence term in Eq. (5.7) has zero integral over all space and, using conventional language, represents the transfer of $\overline{C'^2}$ from place to place. The last term on the right-hand side of Eq. (5.7) constitutes a drain for $\overline{C'^2}$ and can be associated with a dissipation rate of the quantity $\overline{C'^2}$. A resemblance of Eq. (5.7) to the equation for turbulent kinetic energy is evident; only an advection term ($\underline{v} \cdot \nabla \overline{C'^2}$) is missing as a consequence of the fact that reference is made to the moving coordinate system, in which $\underline{v} = 0$.

Immediately after the deployment of, say a Gaussian cloud, the concentration distribution $C(x,t)$ resembles that of the initial distribution $C(x,0)$, and since $\overline{C^2} = \bar{C}^2 + \overline{C'^2}$, the ratio $\overline{C'^2}/\bar{C}^2 \approx 0$ in this limit for small t . In the limit for large times, on the other hand, Chatwin and Sullivan show that, as a consequence almost entirely of molecular diffusion (present through the dissipation rate in Eq. (5.6)), the magnitudes of \bar{C} and $\overline{C'^2}$ decay to zero in a way which depends on the details of the fine-scale

structure of the velocity field. This is probably one reason why experimental measurement of diffusion of gases and heat show that $\overline{C'^2}$ remains of the same order as \overline{C}^2 as plume or clouds develop.

Disregarding, for a while, the molecular diffusivity κ in Eq. (5.5), it is seen that the statistical theory derived in Chapter 3 is inconsistent with the Eulerian fluid description, when C' and thereby $\nabla \cdot (\underline{v}'C')$ is not zero. Therefore, the statistical theory leading to Eq. (3.36) becomes consistent with the fluid description only, when a time-dependent eddy diffusivity $1/2 \, d\sigma^2/dt$ is used to model the flux term $\overline{\underline{v}'C'} = 1/2 \, d\sigma^2/dt \, \nabla \overline{C}$.

In order to experimentally verify the derived formula for relative diffusion (Eq. (3.36)), the two-particle covariance function $R_{abs}(\xi, \tau)$, or its corresponding spectrum function $S(k, \omega)$, has to be estimated from the turbulent field in question. This is especially so when the travel time t is of the same order of magnitude as the integral time scale t_L . From a practical point of view, however, this is rather inconvenient, because reliable Lagrangian statistics of a flow-field are difficult, if not impossible, to obtain. Hay and Pasquill (1959) proposed a working approximation to circumvent this difficulty by assuming that the Eulerian and Lagrangian auto-covariance functions are similar in shape, and that the ratio of the Lagrangian to the Eulerian time scale β is the only parameter to be determined.

Setting $\xi = 0$, this simple hypothesis may be written in the present notation as

$$R_{abs}(0, \beta\tau) = \tilde{R}_{abs}(0, \tau) \quad (5.8)$$

where \tilde{R}_{abs} refers to an Eulerian (fixed point) auto-covariance function.

It will be proposed here that this simple hypothesis applies to the more general situation as well, where the displacement ξ is different from zero, i.e.

$$R_{abs}(\xi, \beta\tau) = \tilde{R}_{abs}(\xi, \tau) \quad (5.9)$$

As also argued by Hay and Pasquill (1959), the assumption on precise similarity between the Lagrangian and Eulerian autocovariance functions is unlikely to produce substantial errors as long as the similarity in shapes are reasonably close.

The relation between the spectrum function $S(k, \omega)$ and its entirely Eulerian spectrum function $\tilde{S}(k, \omega)$ corresponding to it is obtained by simply substituting $\tau = \beta t$ in Eq. (3.40). Then the spectrum can be written:

$$S(k, \omega) = \beta \tilde{S}(k, \beta\omega) \quad (5.10)$$

This shows that the shape of the spectrum function $S(k, \omega)$ and the entirely Eulerian spectrum function $\tilde{S}(k, \omega)$ are also found to be similar. In close analogy with Hay and Pasquill's working approximation, Eq. (5.10) implies that the value of the spectrum function S , at a fixed value of k and at the frequency ω , is equal to the Eulerian spectrum function \tilde{S} at wavenumber k , and at frequency $\beta\omega$.

An alternative to direct measurements of the covariance function $R_{abs}(\xi, \tau)$, namely Taylor's suggestion Eq. (1.2), has already been analysed in Section 3.3.

In their study, Smith and Hay (1961) consider the growth of a Gaussian cloud in a three-dimensional, isotropic field of turbulence. However, the covariance function $R_{abs}(\xi, \tau)$ appears as an entirely Eulerian covariance function $\rho_E(\xi + \bar{u}\tau)$ as a consequence of the following simplifying assumptions: 1) The cloud is assumed to expand in a "quasi-stationary" manner, whereby the relative velocity covariance function $R_{rel}(t, \tau) \approx R_{rel}(\tau)$, is a function of the time lag, τ , alone. 2) The Lagrangian and Eulerian covariance functions, $R_{rel}(\tau)$ and $\tilde{R}_{rel}(\tau)$, respectively, are assumed to be similar in shape, the ratio of the respective time scales being β .

In a recent study, Van Buijtenen (1982) proposes two methods to express the mixed space-time covariance function $R_{abs}(\xi, \tau)$ as a function of an Eulerian space covariance function and a time covariance function: the first is based on a statistical consideration and the other on the basis of physical analogy with mixed longitudinal and lateral space correlations. The statistical approach seems to be the more general and useful; the second approach, however, is simpler and can be useful in specific cases.

The above-mentioned methods, all designed to circumvent the difficulty associated with a direct measurement of $R_{abs}(\xi, \tau)$ from the turbulent field in question, seem to have in common that they suffer from a lack of experimental verification.

6. CONCLUSIONS

A statistical theory for the turbulent dispersion of a Gaussian cloud has been proposed in terms of the two-particle covariance function $R_{abs}(\xi, \tau)$ (cf. Eq. (3.36) or its equivalent Fourier transform Eq. (3.47) or (3.62)).

In order to do so, the following assumptions have been introduced:

- 1) The turbulent field is taken to be homogeneous, stationary, and one-dimensional.
- 2) Equivalence has been assumed between ensemble-averaged concentration functions and probability distribution functions.
- 3) The nature of the individual fluid particles displacement process Δy_i , in the clouds centre-of-mass coordinate system is assumed to follow identical and independent Gaussian statistics, cf. Eq. (3.19).

The last of these assumptions is the most fundamental for the present theory, and the instantaneous Gaussian cloud shape results as a consequence hereof.

Applicable for diffusion times that are small compared with the integral time scale of the turbulence, simple expressions for the growth of the standard deviation $\sigma(t)$ of the puff have been derived by assuming that the wavenumber spectrum corresponding to the Eulerian space covariance is a power law δk^p .

For the inertial subrange in atmospheric turbulence, where $p = -5/3$, the predictions (Eq. (3.69)) of the cloud growth are found to be consistent with Batchelor's (1950) similarity theory, both at "small" and at "intermediate" times. In addition to the results of similarity theory, the constant of proportionality between σ^2 and ϵt^3 has also been calculated to be 0.534 (see Eq. (4.2)).

For the case of inertial range two-dimensional turbulence, where $p = -3$, the theory predicts exponential growth in agreement with dimensional analysis by Lin (1972).

Finally, a simple working approximation, Eq. (5.9), is suggested for the determination of the covariance function $R_{abs}(\xi, \tau)$ in terms of entirely Eulerian fields.

ACKNOWLEDGEMENTS

The author is indebted to Prof. C.M. Tchen, City University of New York, for many useful discussions. Also my colleagues Ib Troen, Søren Larsen, Hans Pécseli, and Leif Kristensen are gratefully acknowledged for their interest and suggestions.

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APPENDIX A

Important notation

<u> </u>	ensemble average over all realizations of turbulent field
< >	average over particles in cluster = $1/N \sum_{i=1}^N$
'	deviation from cluster mean
<u>a</u>	vector quantity of magnitude a
c	centre-of-mass position of the cloud, and origin of the moving frame co- ordinate, y
C	concentration distribution function of cloud
d	linear extension of a fluid particle
G	Gaussian particle distribution function
H	Heavyside's step function
i	(as suffix) designation of individual particles (x_i) or fluid element (dx_i) in the cloud
k	wavenumber ($= 2\pi/\lambda$ where λ is wave- length
K_R	eddy diffusivity for relative diffusion

K	eddy diffusivity for absolute diffusion
ℓ	distance separating two typical marked fluid elements
ℓ_E	Eulerian integral length scale
M	number of fluid particles in the turbulent field
N	number of tracer particles in a cloud
n	difference in particle number $i-j$
p	exponent in power function k^p
$P(x', x'', t)$	two-particle displacement probability density (see Eq. (2.14))
q	distance neighbour function (see Eq. (1.2))
Q	total amount of matter released with a cloud
r	relative velocity correlation function (see Eq. (2.22))
$R_{abs}(\tau)$	Lagrangian auto-covariance function, $\frac{u(t)u(t-\tau)}{}$
$R_{abs}(\xi, \tau)$	two-particle velocity covariance function (see Eq. (3.26)). (Note that $R_{abs}(0, \tau) \equiv R_{abs}(\tau)$)
$R_{cm}(t, \tau)$	centre-of-mass covariance function (see Eq. (3.6))

$R_{rel}(t, \tau)$	relative velocity covariance function (see Eq. (3.6))(Note $R_{rel}(t, \tau) = \overline{r(t, \tau)^2}(\tau)$)
s	standard deviation for the relative displacement process Δy_i (see Eq. (3.18))
$S(k, \omega)$	spectrum function corresponding to covariance $R_{abs}(\xi, \tau)$
$S_E(k)$	spectrum function corresponding to $\rho_E(\xi)$
$S_L(\omega)$	spectrum function corresponding to $\rho_L(\tau)$
t	time, with origin at moment of release of cloud
t_E	fixed frame (Eulerian) integral time scale
t_L	fixed frame (Lagrangian) integral time scale
$t_r(t)$	Lagrangian integral time scale appropriate for relative diffusion (see Eq. (2.23))
T_c	time scale $\eta^{-1/3}$ in the enstrophy inertial subrange
u	velocity component referred to fixed frame (x)
v	velocity component referred to moving frame (y)

V_{cm}	centre-of-mass velocity component dc/dt
x	fixed or absolute frame coordinate
y	moving or relative frame coordinate
Δy_i	relative frame displacement process (see also Fig. 3)
α	dimensionless constant of order unity (see Eq. (4.1))
β	ratio of Lagrangian to Eulerian integral time scales, viz. t_L/t_E
$\Gamma(p)$	gamma function $\int_0^\infty x^{p-1} e^{-x} dx$
Δ	$(\overline{u^2 t_L^2})^{1/2}$ (see Eq. (4.11))
δ	coefficient to power law $S_E(k) = \delta k^p$ with dimension $m^{(1+p)}$
ϵ	rate of dissipation of energy
η	enstrophy cascade rate
κ	molecular diffusivity
μ	fourth moment of the concentration distribution about the centre of mass
ν	kinematic viscosity
$\xi_{ij}(t)$	separation of two particles i, j at fixed time (see Eq. (3.22))
$\rho_E(\xi)$	Eulerian correlation coefficient $R_{abs}(\xi, 0)/\overline{u^2}$

$\rho_L(\tau)$	Lagrangian correlation coefficient $R_{abs}(0, \tau) / \overline{u^2}$
σ	standard deviation of the particle positions about the centre of the Gaussian puffs
σ_0	initial puff size $\sigma(t = 0)$
τ	time lag (see Fig. 1)
ω	angular frequency ($= 2\pi \times$ cycles per unit time)

APPENDIX B

Spectral definitions

This appendix justifies some of the spectral relations used in the body of the report.

The spectrum of the two-particle, mixed Lagrangian-Eulerian covariance function $R_{abs}(\xi, \tau)$ is defined through the Fourier transform pair

$$S(k, \omega) = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} R_{abs}(\xi, \tau) \exp(-i(k\xi + \omega\tau)) d\xi d\tau \quad (B.1)$$

$$R_{abs}(\xi, \tau) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \exp(i(k\xi + \omega\tau)) dk d\omega \quad (B.2)$$

By definition, $R_{abs}(0, 0) = \overline{u^2}$, so from Eq. (B.2) with $(\xi, \tau) = (0, 0)$ we have

$$\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) dk d\omega = \overline{u^2} \quad (B.3)$$

For the case where $\xi = 0$, which corresponds to an entirely Lagrangian (single-particle) correlation function $\rho_L(\tau) = \overline{u(x, t) u(x, t - \tau) / u^2}$, we also define the Fourier transform pair

$$S_L(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \rho_L(\tau) \exp(-i\omega\tau) d\tau \quad (B.4)$$

$$\rho_L(\tau) = \int_{-\infty}^{\infty} S_L(\omega) \exp(i\omega\tau) d\omega \quad (B.5)$$

Since $\rho_L(0) = 1$, we have from Eq. (B.5)

$$\int_{-\infty}^{\infty} S_L(\omega) d\omega = 1$$

Analogously, for the case where $\tau = 0$, which corresponds to an entirely Eulerian two-point correlation function $\rho_E(\xi) = \overline{u(x,t)u(x+\xi,t)/u^2}$ we define the Fourier transform pair

$$S_E(k) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \rho_E(\xi) \exp(-ik\xi) d\xi \quad (B.6)$$

$$\rho_E(\xi) = \int_{-\infty}^{\infty} S_E(k) \exp(ik\xi) dk \quad (B.7)$$

Since $\rho_E(0) = 1$, we have from Eq. (B.7)

$$\int_{-\infty}^{\infty} S_E(k) dk = 1 \quad (B.8)$$

By noting that $R_{abs}(0, \tau) = \rho_L(\tau) \overline{u^2}$, we get from Eq. (B.2), by setting $\xi = 0$

$$\overline{u^2} \rho_L(\tau) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \exp(i\omega\tau) dk d\omega \quad (B.9)$$

A comparison with Eq. (B.4) now shows the relation

$$\overline{u^2} S_L(\omega) = \int_{-\infty}^{\infty} S(k, \omega) dk \quad (B.10)$$

Analogously, by noting that $R_{abs}(\xi, 0) = \rho_E(\xi) \overline{u^2}$, we get from Eq. (B.2) with $\tau = 0$, and Eq. (B.7), the relation

$$\overline{u^2} S_E(k) = \int_{-\infty}^{\infty} S(k, \omega) d\omega .$$

